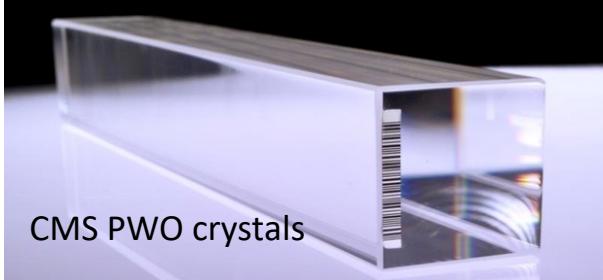


“Quantum” scintillator materials

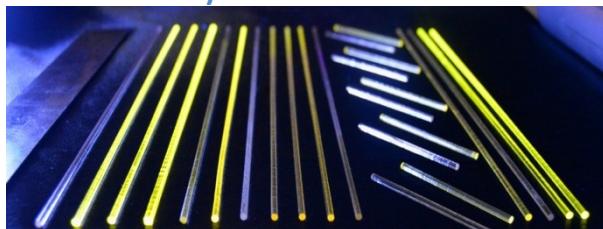
E. Auffray, *CERN, EP-CMX*
Crystal Clear (RD18) Collaboration Spokesperson

“Standard” Scintillators

Bulk crystals

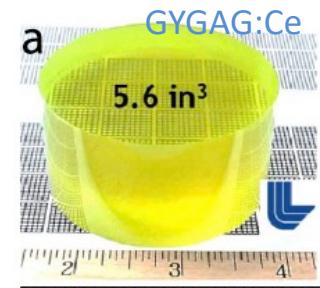


Crystal fibers

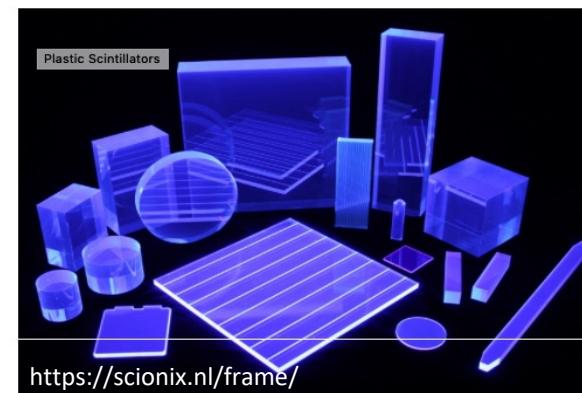


Several types of crystal Fibers from ILM;
Courtesy K. lebbou, ILM Lyon

Ceramics



Plastic scintillators

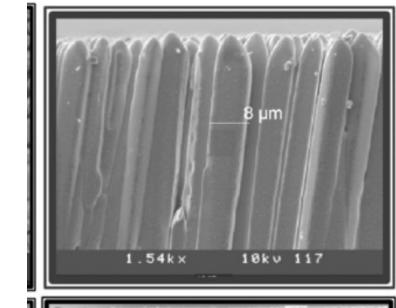


Thin film



J. Tous et al. ,Radiation Measurements
47 (2012) 311-314

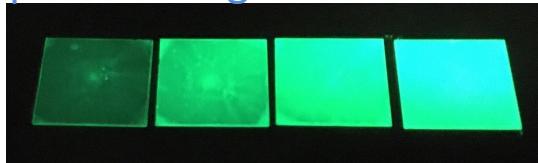
CsI(Tl) needles



S. Miller et al. August 2005
DOI: [10.1111/j.1365-2729.2005.00927.x](https://doi.org/10.1111/j.1365-2729.2005.00927.x)

Next generation: “quantum” scintillators ?

CsPbBr₃ Nano crystals thin films
deposited on glass substrate



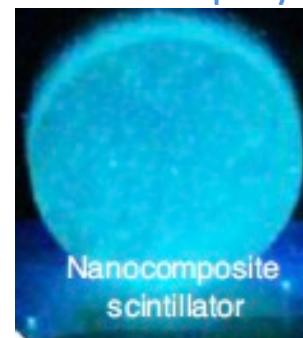
Courtesy V. Čuba, K. Děcká, A. Suchá CTU, Prague

ZnO:Ga nanocrystals
embedded in SiO₂



L. Procházková et al., Radiat Meas 90, 2016, 59-63
R. Turto Phys. Status Solidi RRL 10, No. 11, 843–847
(2016)

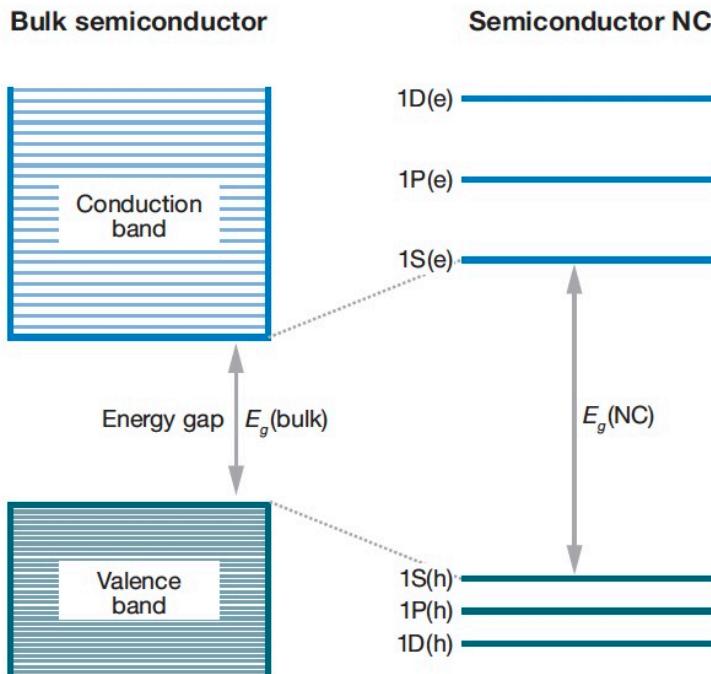
Metal–organic framework (MOF) nanocrystals
Embedded in polymer



J. Perego et al. Nat. Photonics (2021).
<https://doi.org/10.1038/s41566-021-00769-z>

From bulk to nanomaterial: Quantum confinement

Same crystal lattice but nanometer-sized crystal particle



V. Klimov Annu Rev. Phys. Chem. 58 (2007) 535-573

With decreasing crystal size
From “continuous band” to quantized energy levels

Energy band gap (E_g) varies with quantum dot size
Decrease of size => increase of E_g

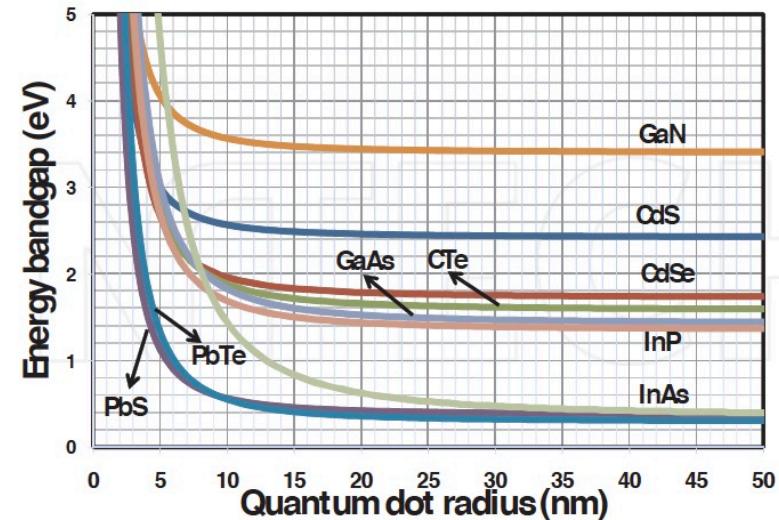
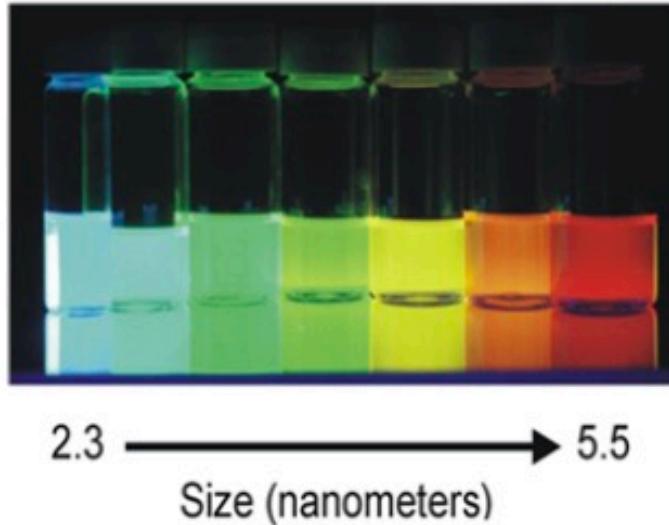


Figure 4. Variation of quantum dot energy bandgap vs. dot size for some common semiconductors. From [9].

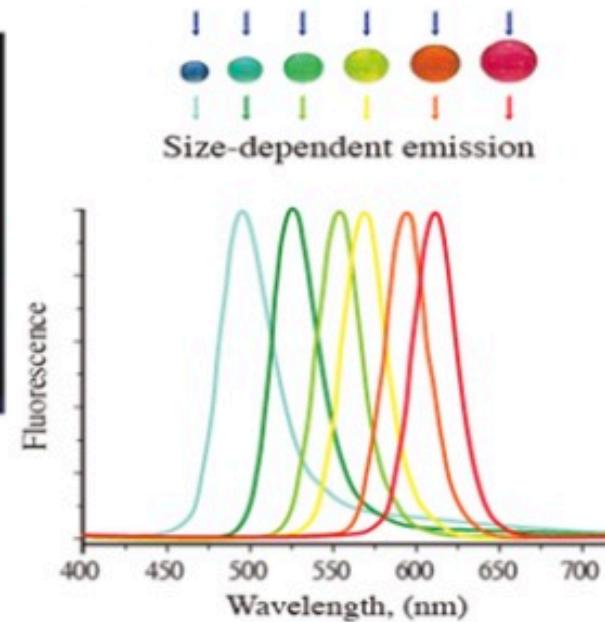
K. Jasim, Quantum Dots Solar Cells <http://dx.doi.org/10.5772/59159>

Quantum confinement

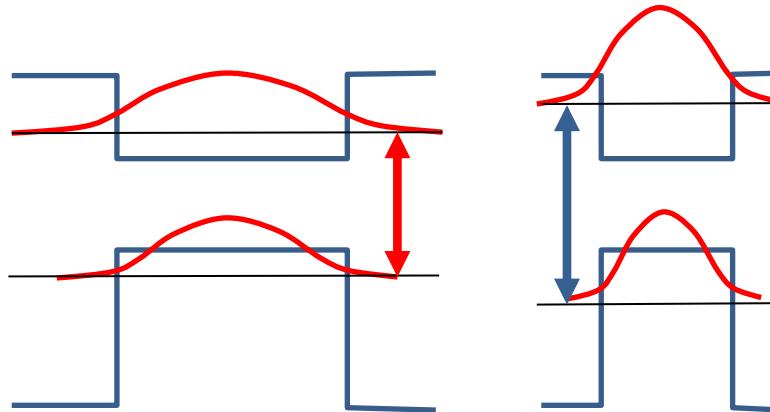
from Benoit Dubertret and Hideki Ooba



Simultaneous excitation at 365 nm



=> Tune the emission properties by changing size of nanodots



Exciton energy increases with decrease of nanostructure size – control of emission wavelength

1,2,3D confinement

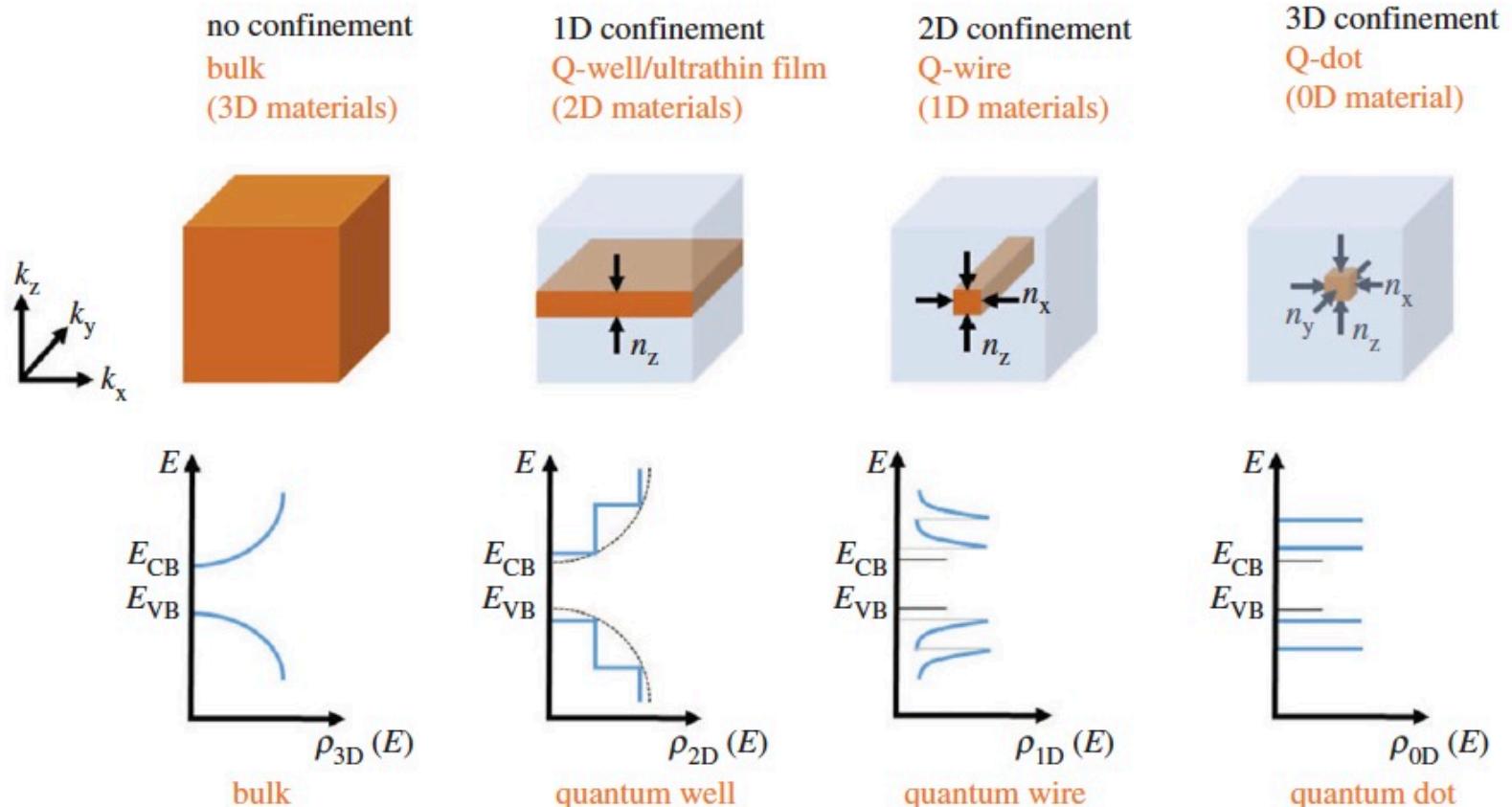
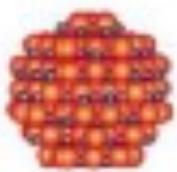


Figure 1. Schematic illustration of broken symmetry and functional form of the density of states in 1D, 2D and 3D confined materials.

5

Example of CdSe

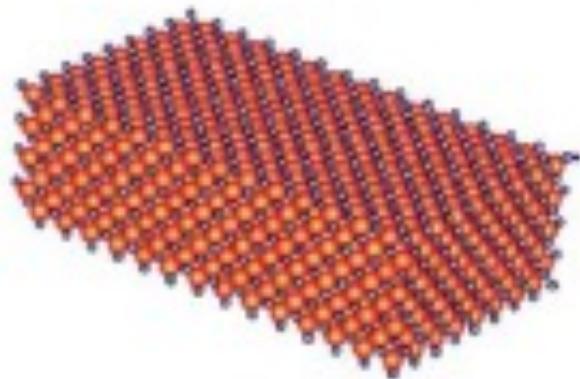
3D Confinement



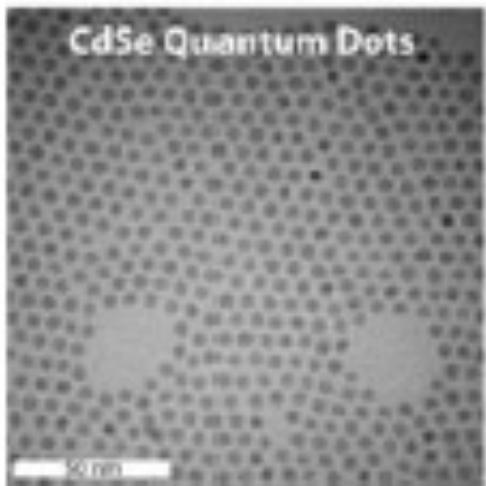
2D Confinement



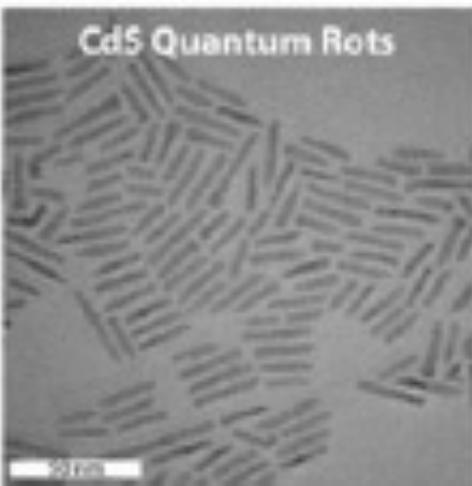
1D Confinement



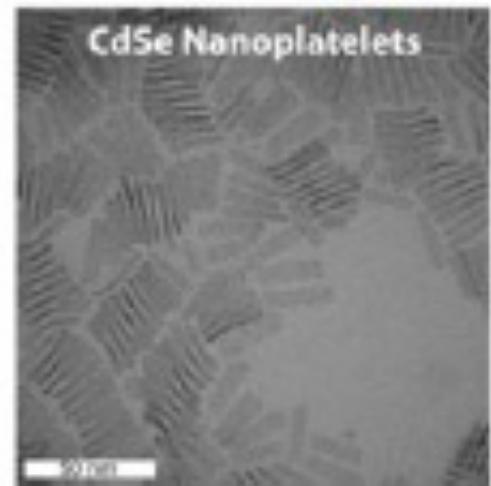
CdSe Quantum Dots



CdS Quantum Rots

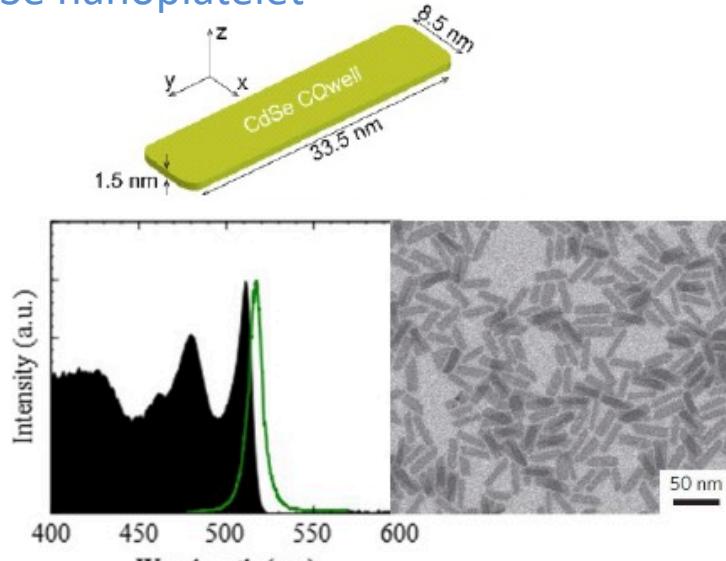


CdSe Nanoplatelets



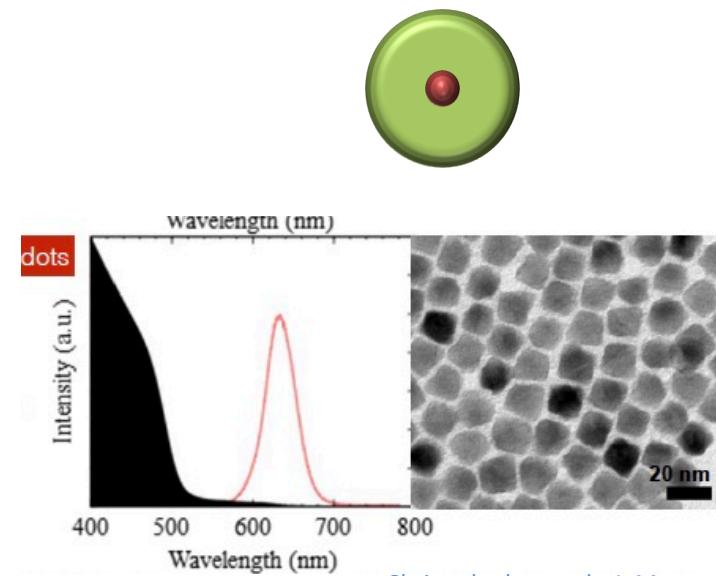
CdSe quantum well/quantum dot

CdSe nanoplatelet



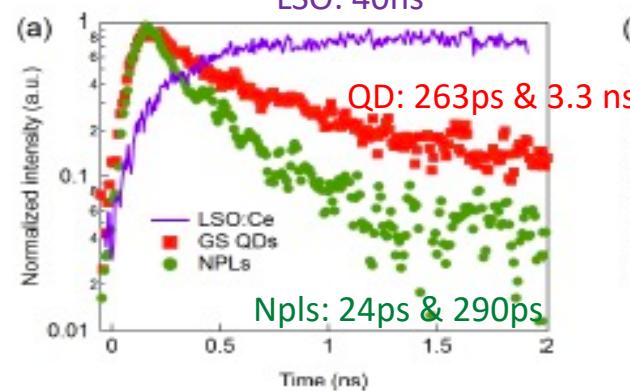
J.Q. Grim et al., Nature Nanotechnol. 9 (2014) 891.

CdSe/CdS Giant shell Quantum dot

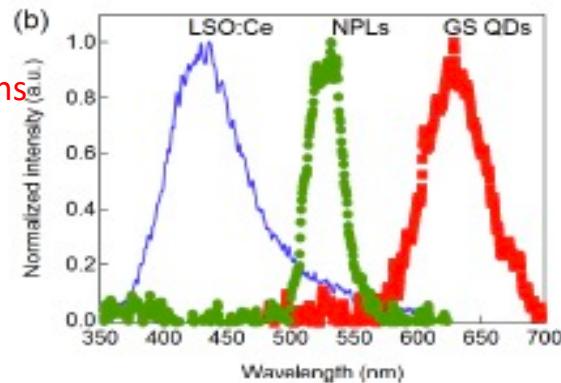


Christodoulou et al., J. Mater. Chem. 2014, 2, 3439.

LSO: 40ns



(b)

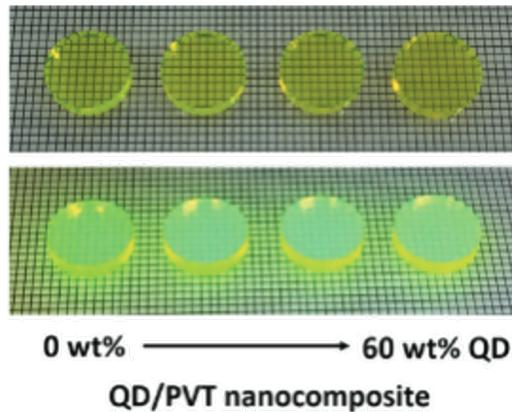


R. Martinez Turtos et al., 2016 JINST_11 (10) P10015

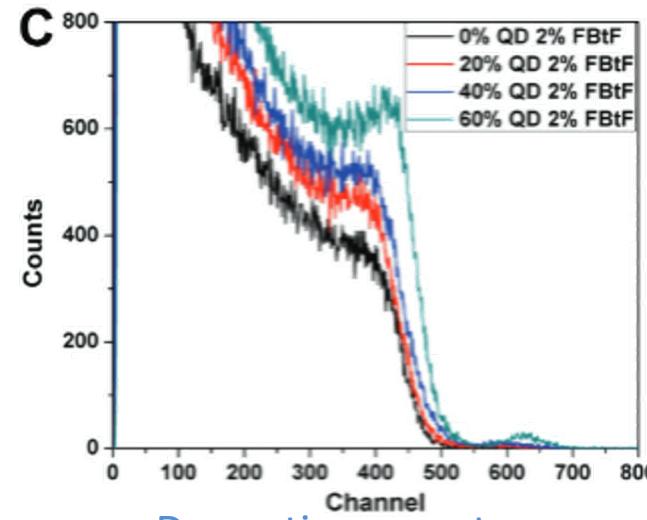
=> Much Faster than LYSO crystal

measured @cern Lab27

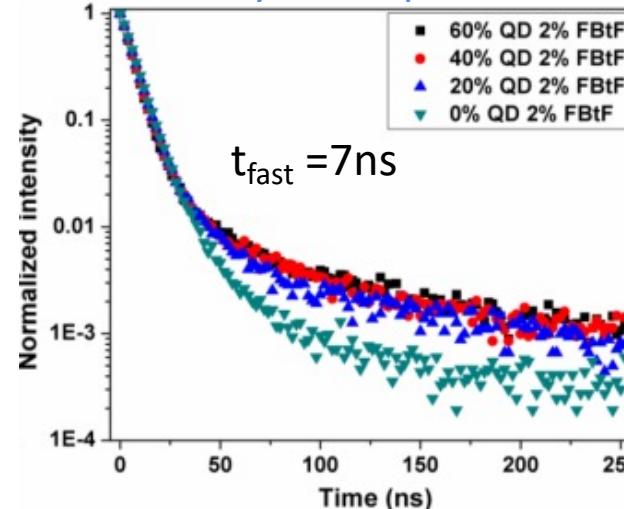
CdxZn_{1-x}S/ZnS (CZS) QD Nanocomposite Synthesis



Cs^{137} photoelectric spectra



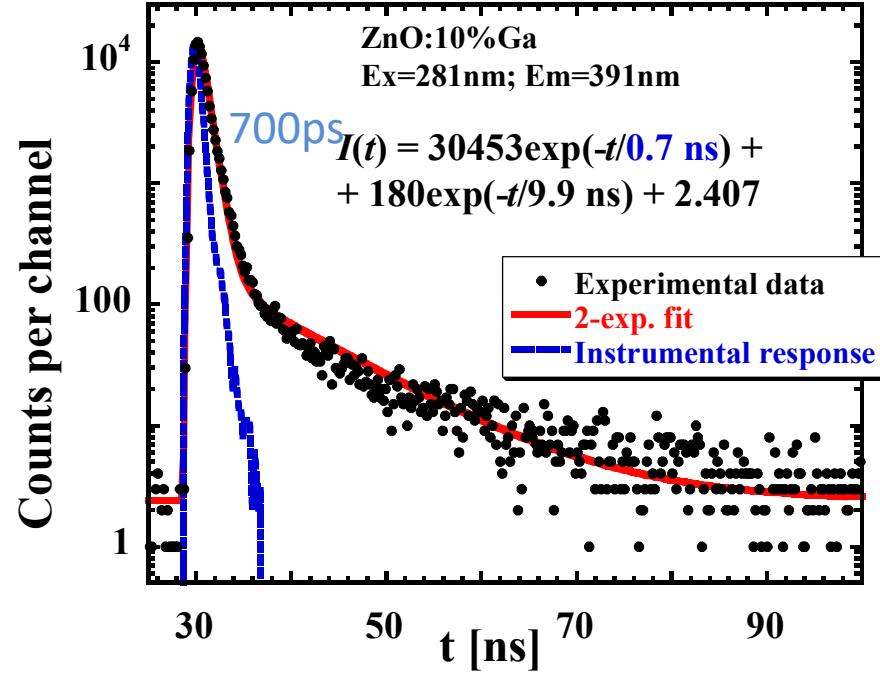
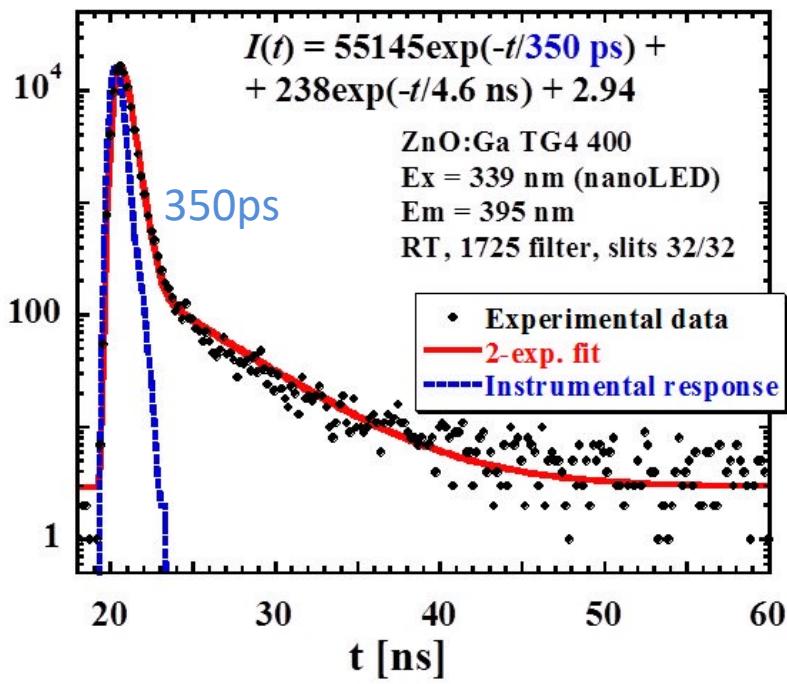
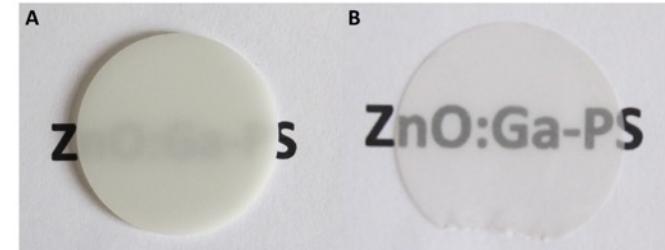
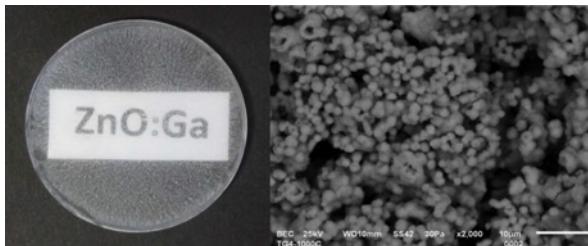
Decay time spectra



ZnO:Ga Nanocomposite: photoluminescence properties

In Polystyrene

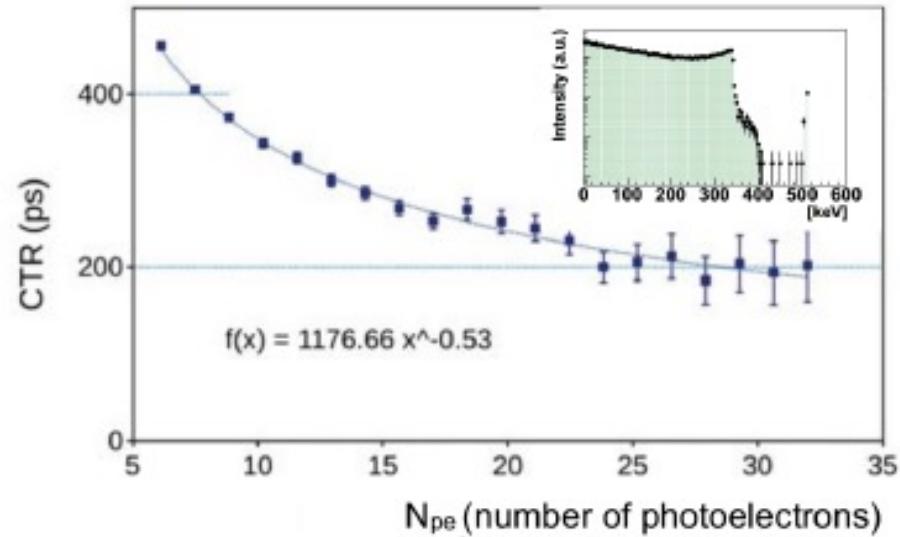
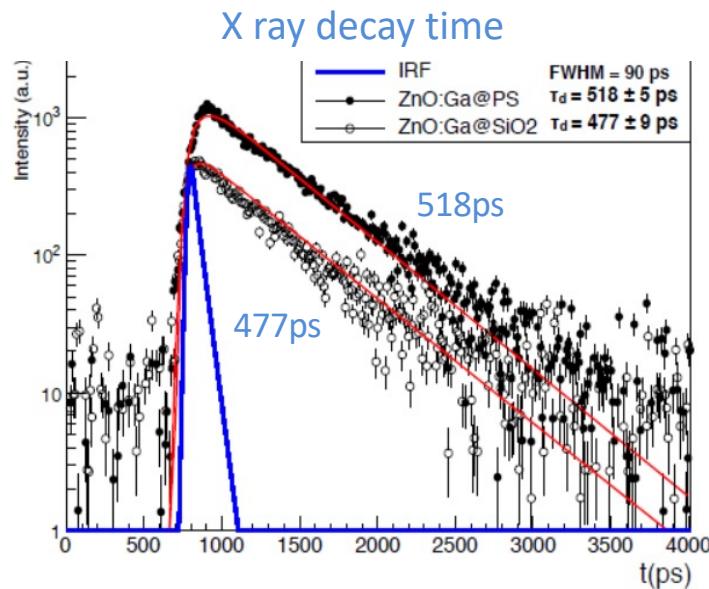
In SiO₂



Procházková et al., Radiat Meas 90, 2016, 59-63

Buresova et al, Opt. Express 24, 15289 (2016)

ZnO:Ga nanocomposite Radioluminescence properties



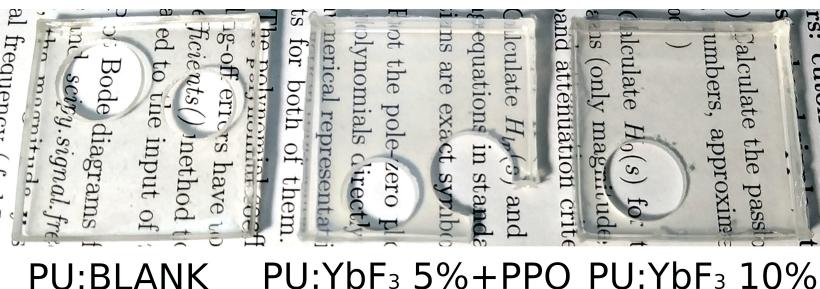
CTR of 200ps with only 30 photons detected by the SiPM due to the very fast scintillation decay
 => With improvement of transparency and reducing self absorption we could get better time resolution



ASCIMAT

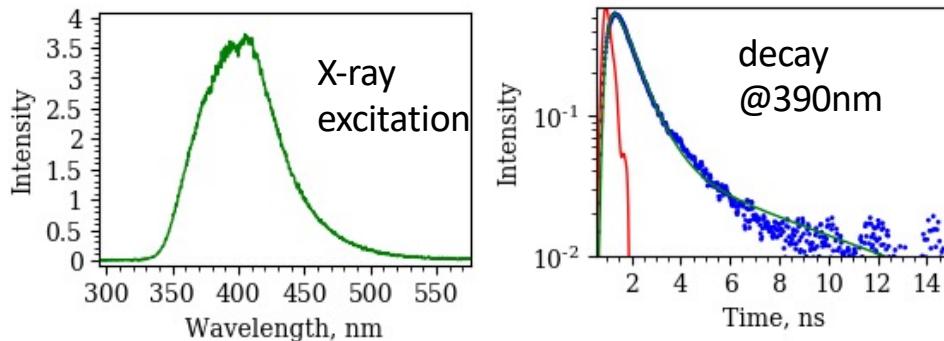
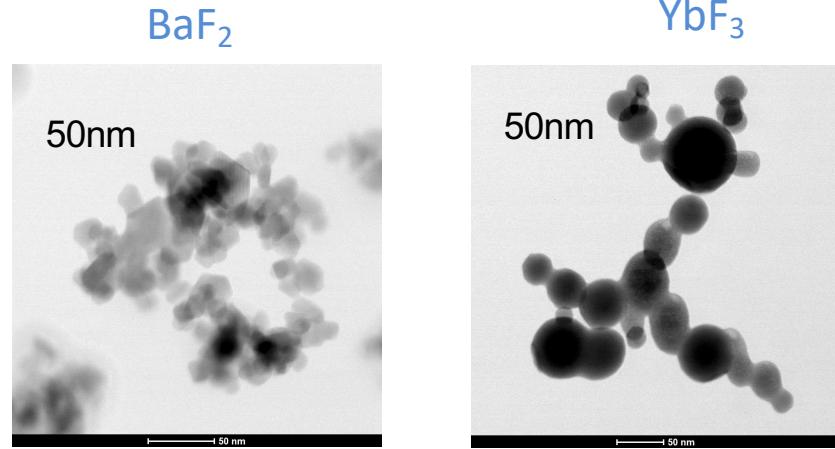
measured @cern Lab27

Polyurethane scintillators filled with BaF₂ and YbF₃ nanocrystals

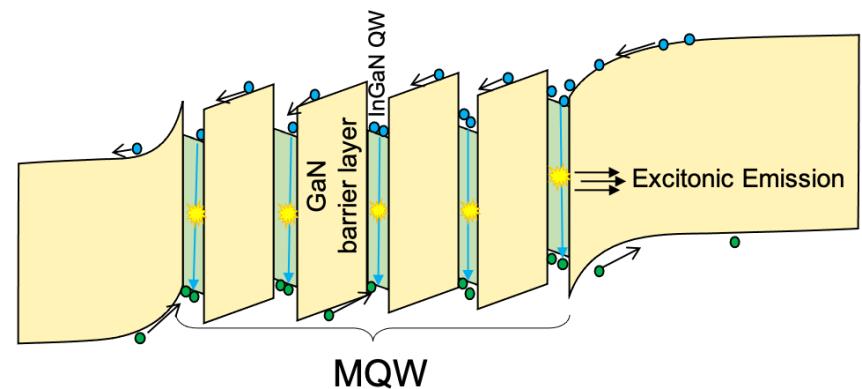
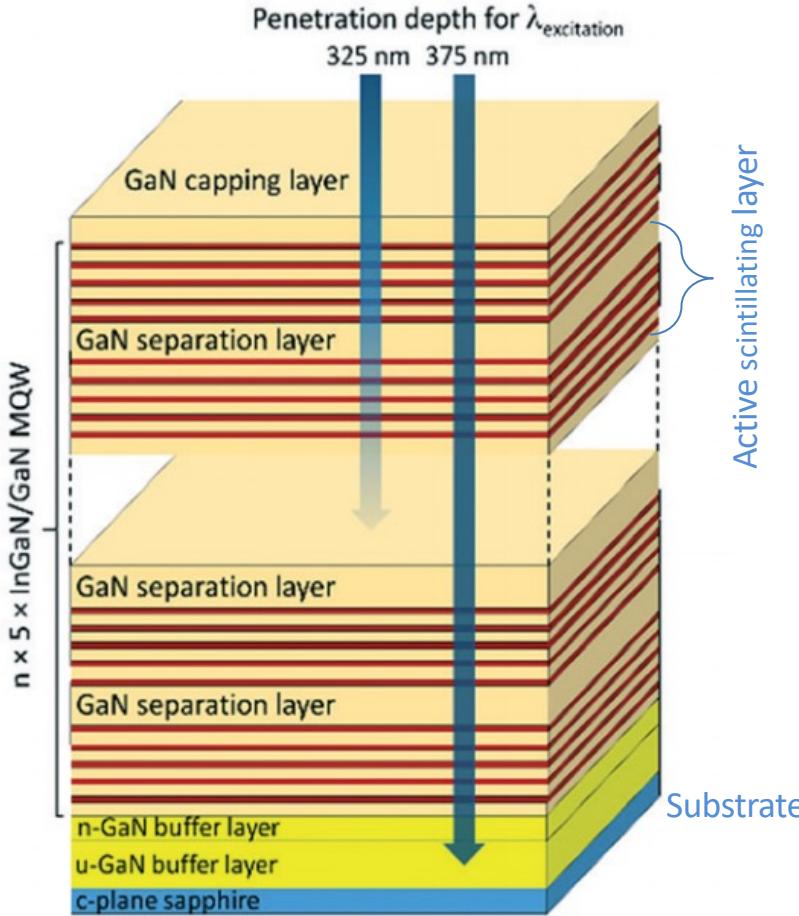


PU:BLANK PU:YbF₃ 5%+PPO PU:YbF₃ 10%

- Two-component polyurethane resin
 - Part A – isocyanate
 - Part B – polyols
- Nanoparticle filler 20-50 nm up to 10wt%
- Cathdoluminescence yield up to 50% of 422 plastic scintillator
- X-ray decay time 0.8 ns
- NO rise time observed



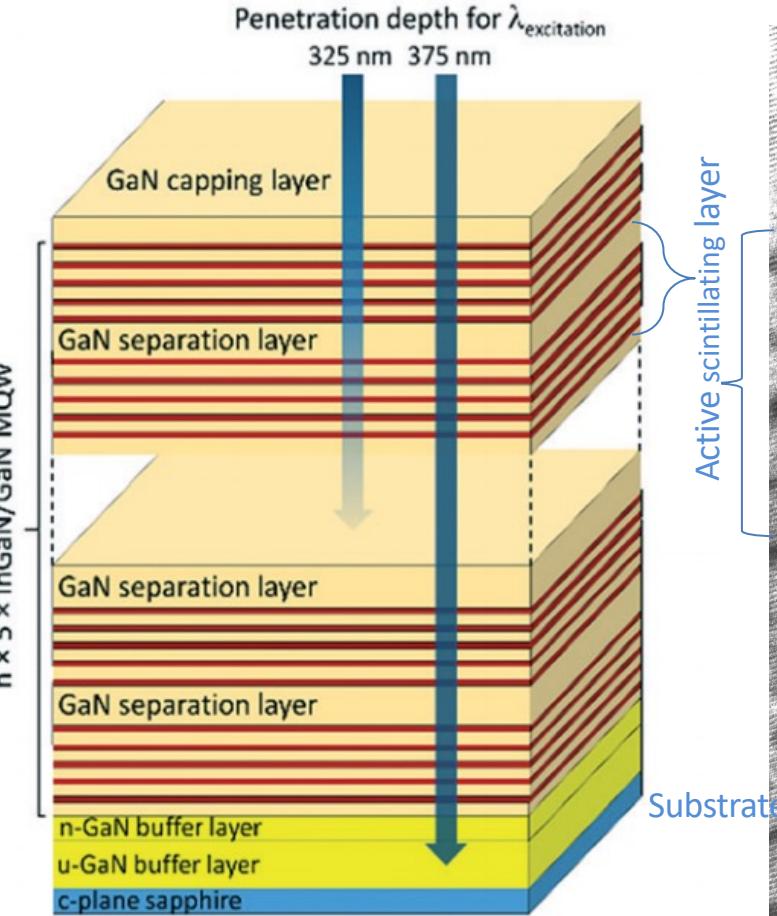
InGaN/GaN heterostructure: Multiple Quantum Wells (MQW)



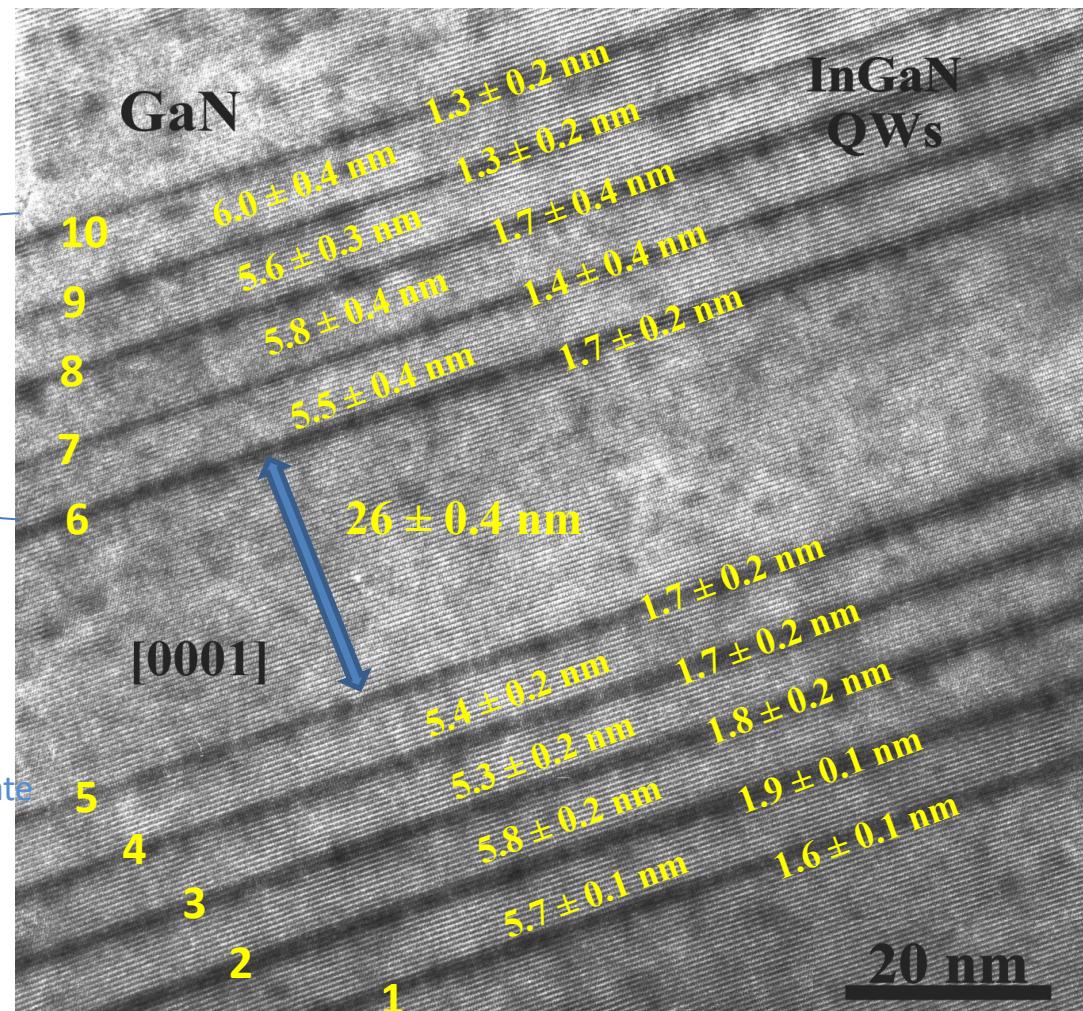
Electrons and holes are concentrated in narrow gap layers and radiatively recombine there being spatially confined by small thickness (few nm) of the layer.

MOVPE technology can prepare such nanostructures on 4-6 inch size Al₂O₃ substrates

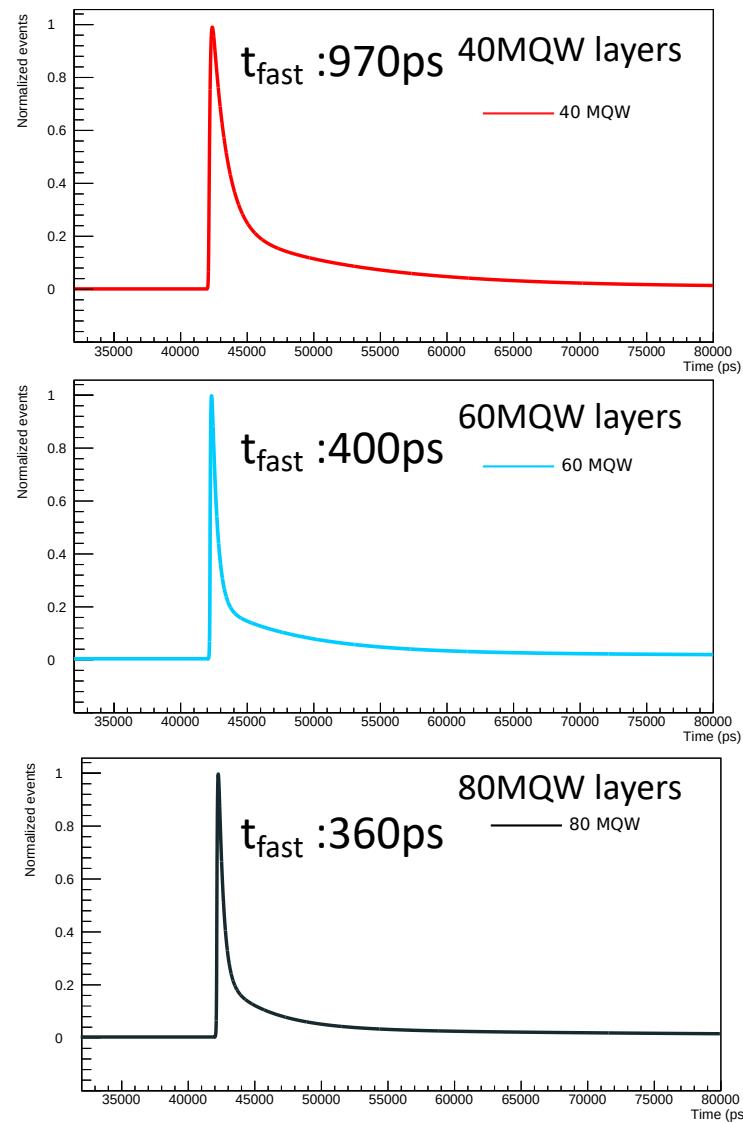
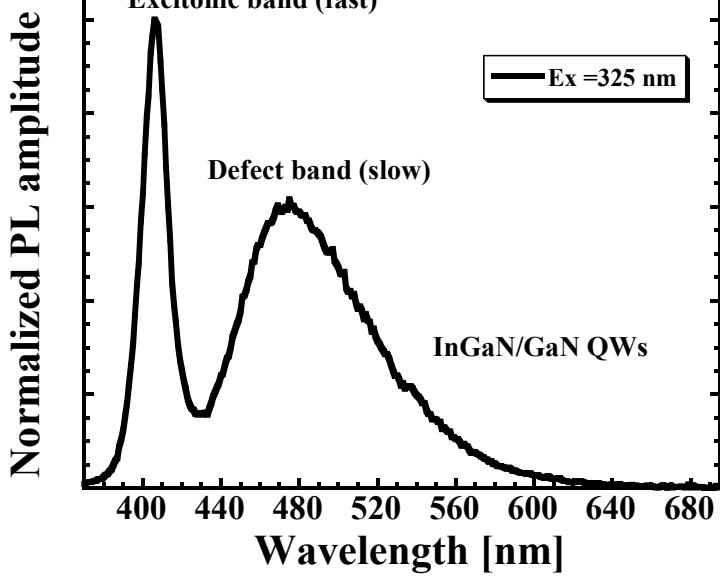
InGaN/GaN heterostructure: Multiple Quantum Wells (MQW)



Picture from A. Hospodkova

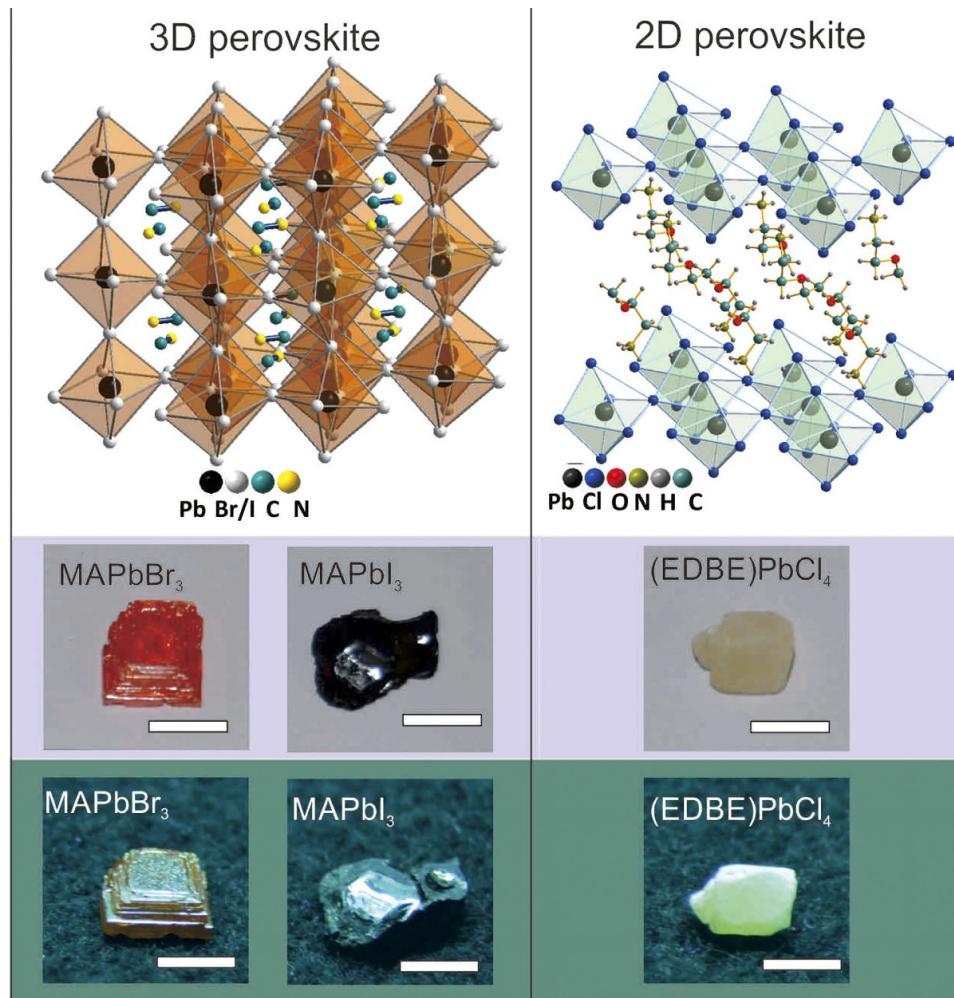


InGaN/GaN heterostructure: Multiple Quantum Wells (MQW)

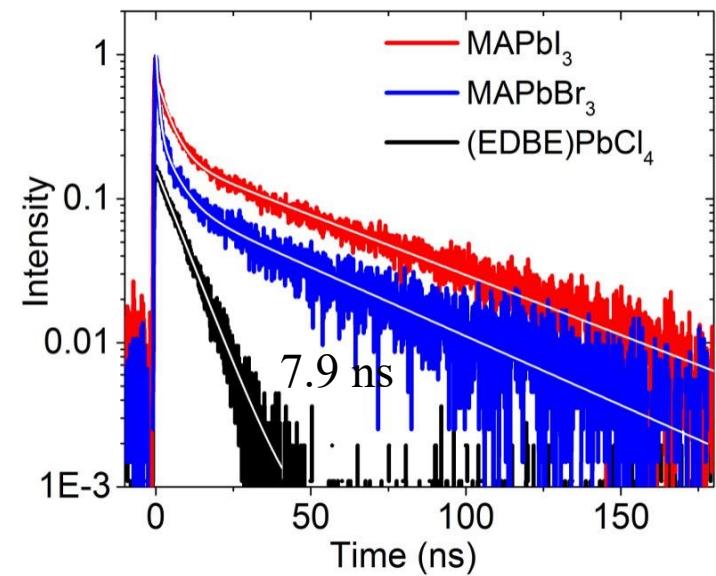


measured @cern Lab27

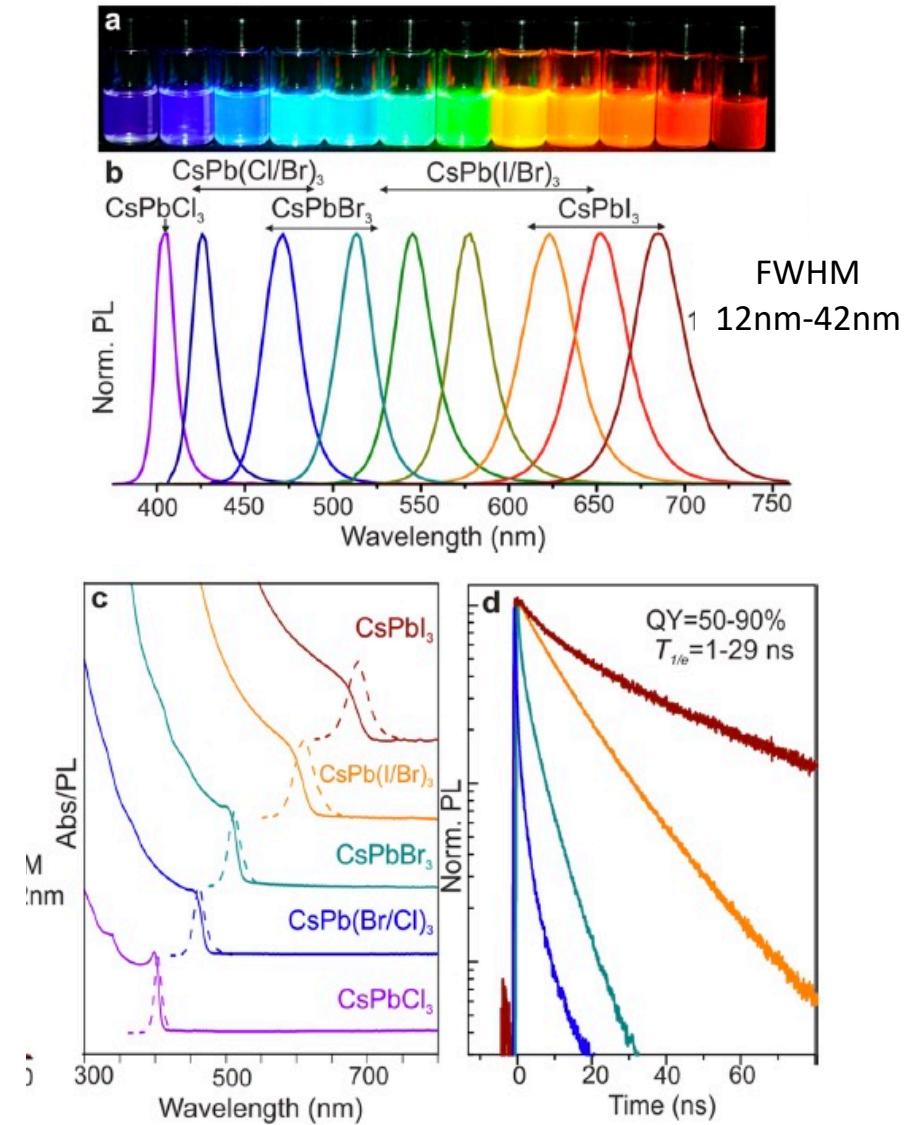
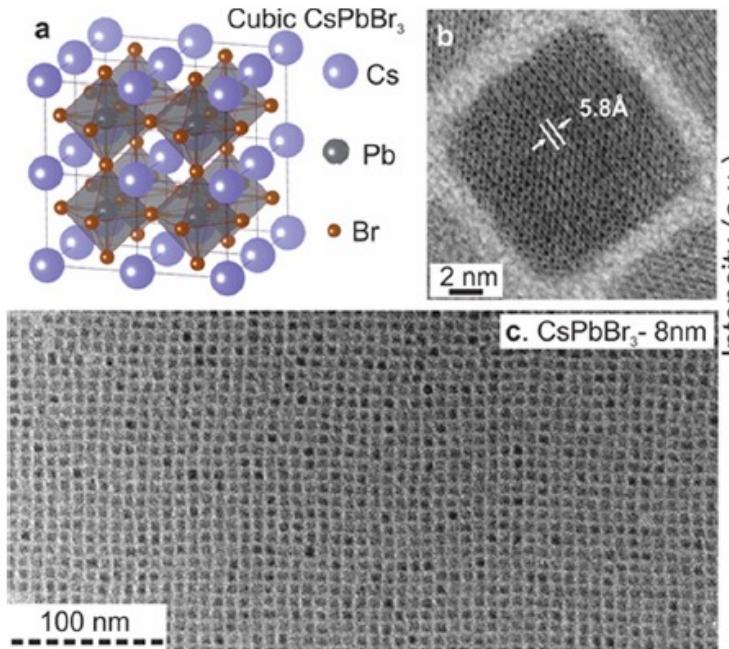
Perovskites material



2D perovskite:
alternating organic and inorganic layers

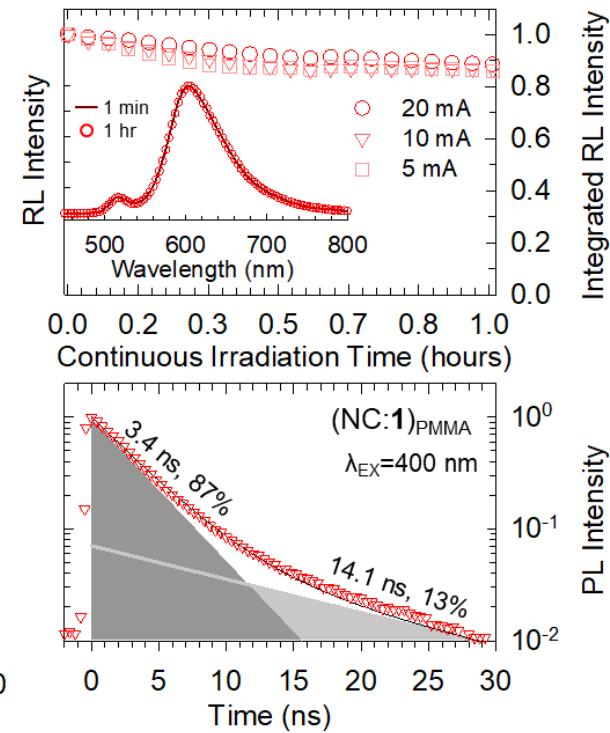
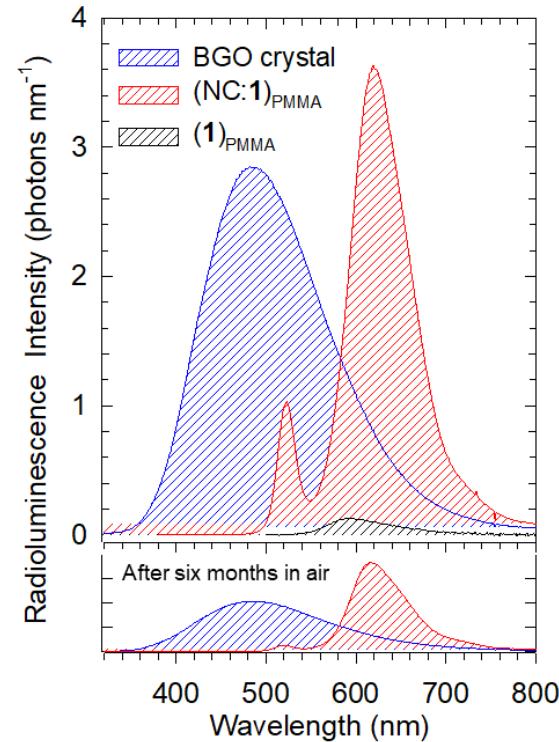
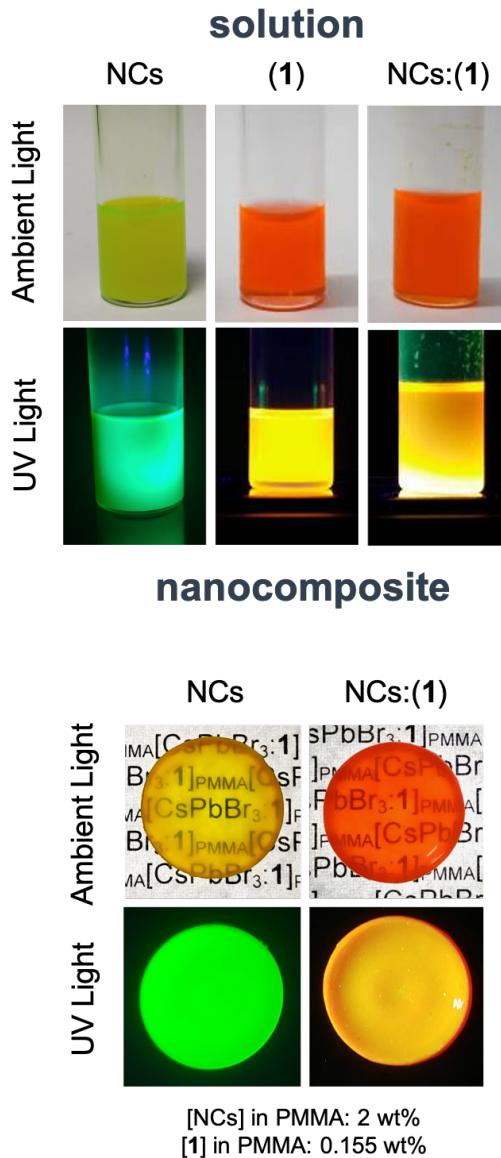


Perovskite nanocrystals



L. Protesescu et al. Nano Lett. 2015, 15, 3692–3696
 DOI: 10.1021/nl5048779

Perovskite nanocrystals



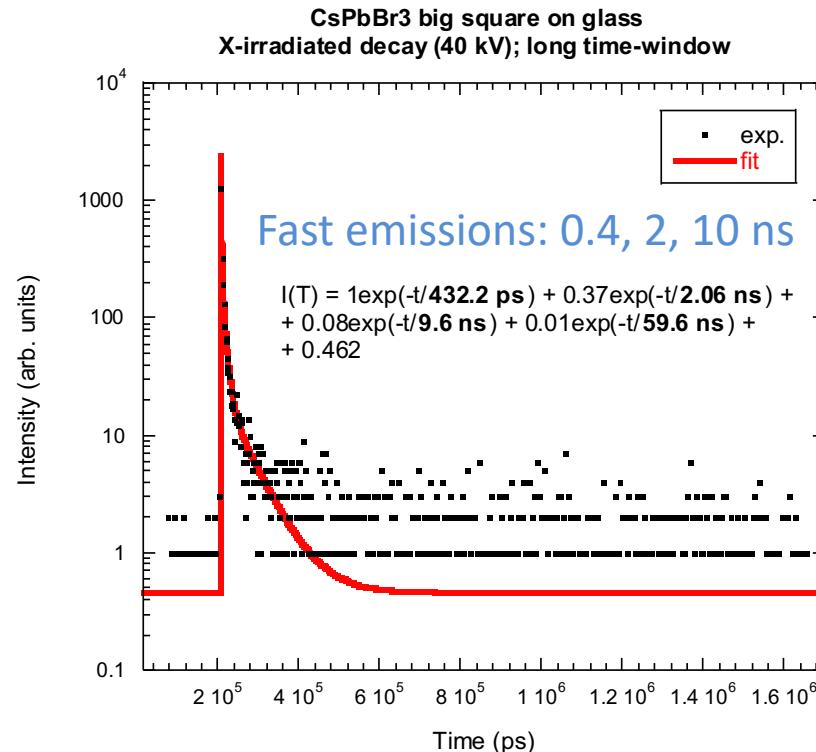
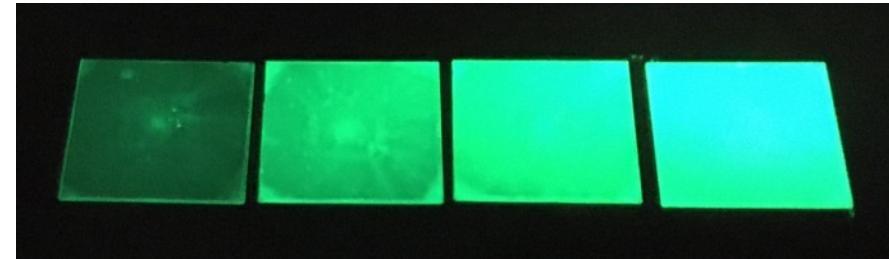
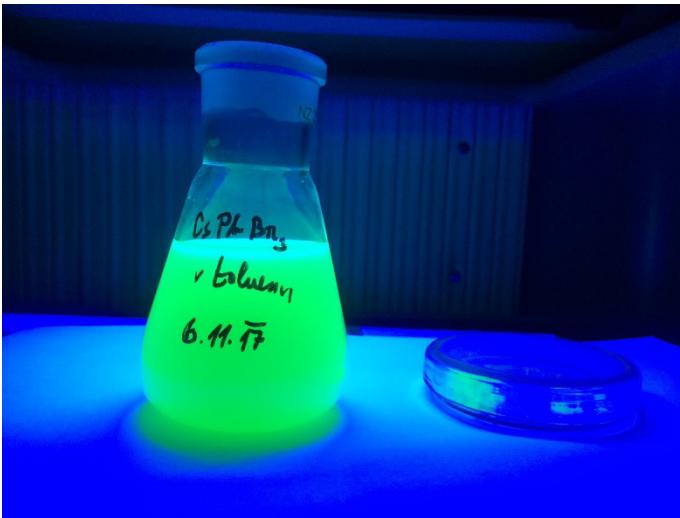
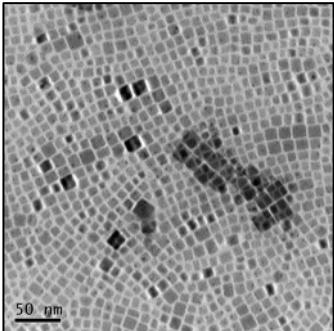
M. Gandini, *Nat. Nanotechnol.* **15**, 462–468 (2020).
<https://doi.org/10.1038/s41565-020-0683-8>



Perovskite thin film

CsPbBr₃ thin films deposited on glass substrate

CsPbBr₃ nanocrystals



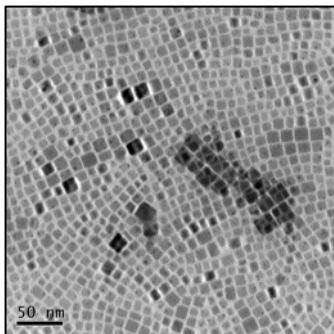
Courtesy V. Čuba, K. Děcká, A. Suchá CTU, Prague



Perovskite nanocomposite



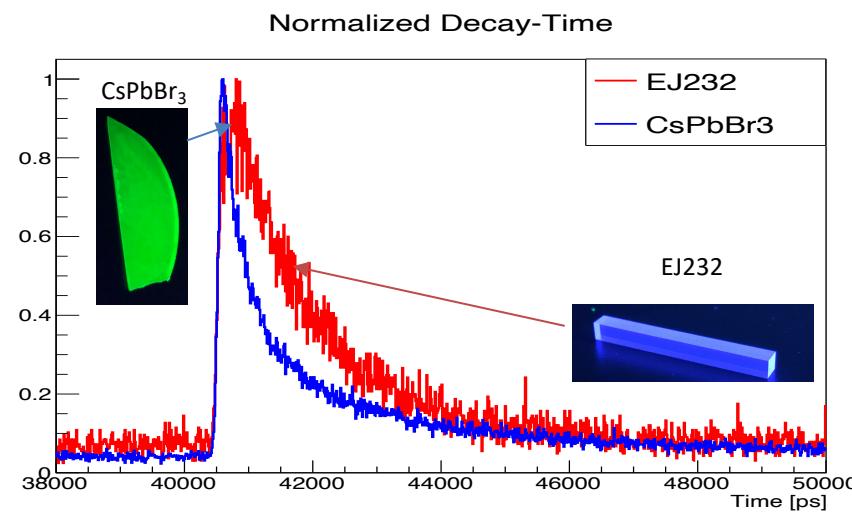
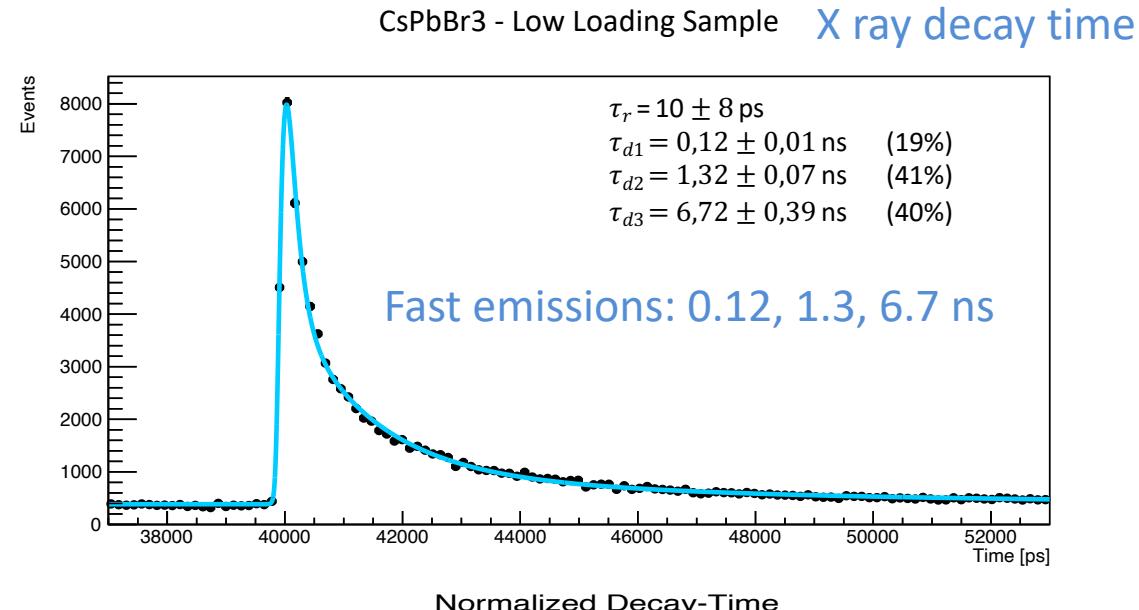
CsPbBr₃ nanocrystals



CsPbBr₃ nanocrystals
imbedded in polystyrene

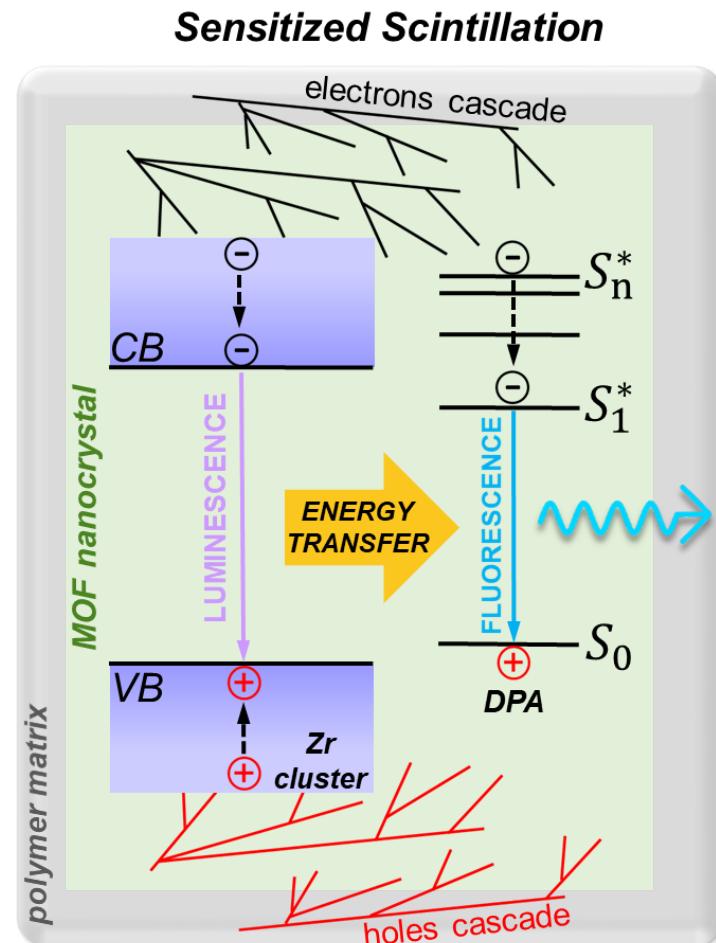
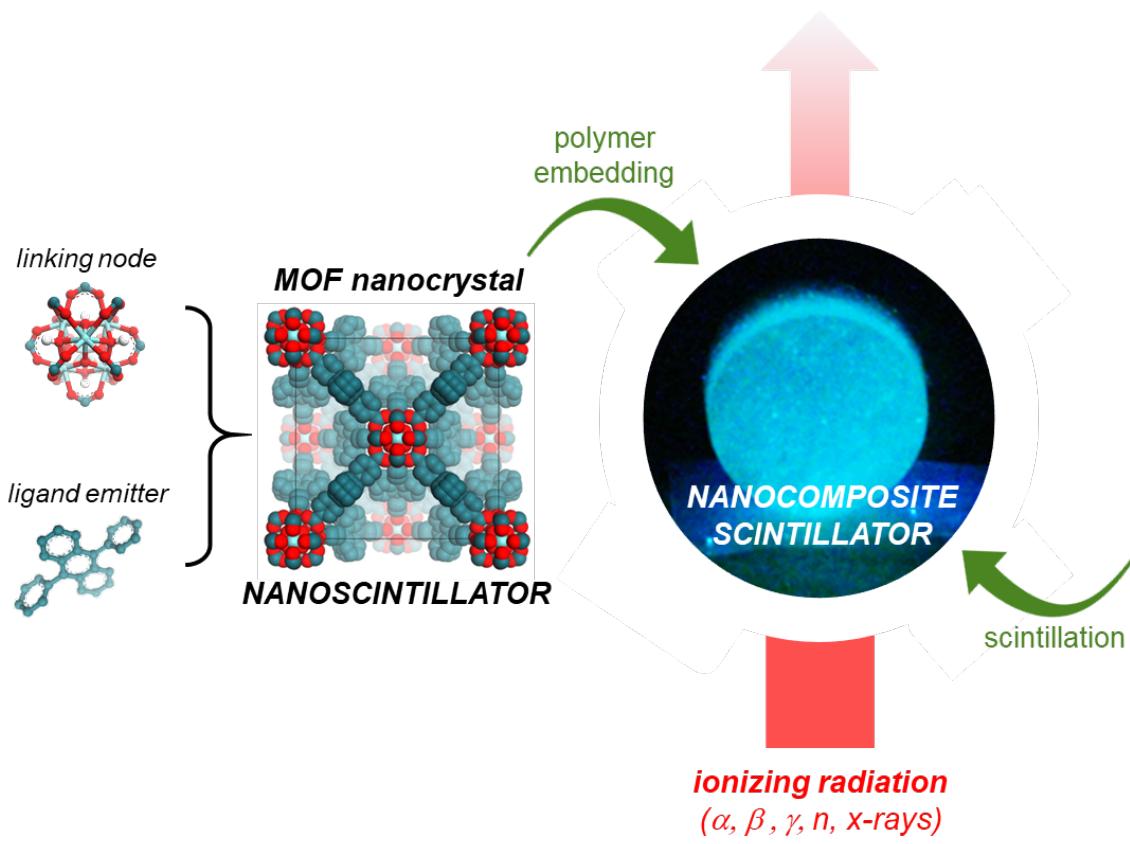


Courtesy V. Čuba, K. Děcká, A. Suchá CTU, Prague

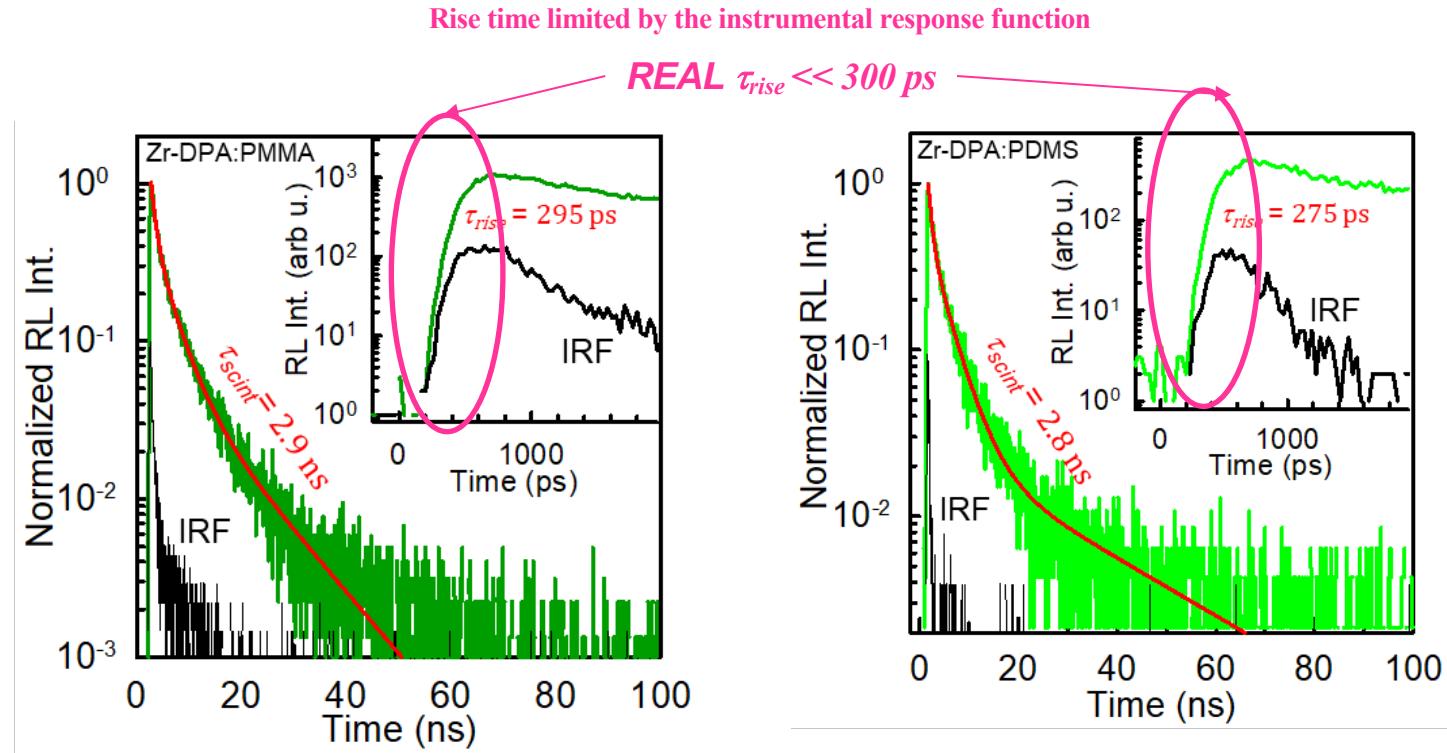


measured @cern Lab27

Composite fast scintillators based on high-Z fluorescent metal–organic framework (MOF) nanocrystals



Composite fast scintillators based on high-Z fluorescent metal-organic framework (MOF) nanocrystals



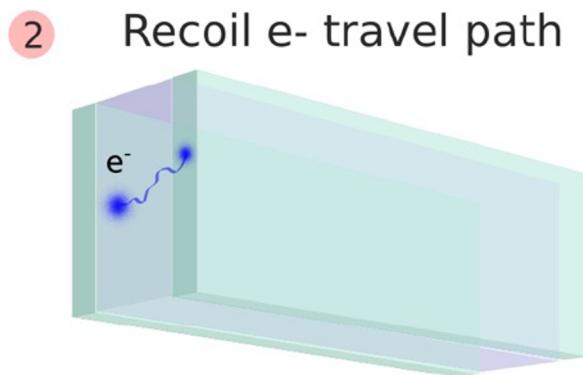
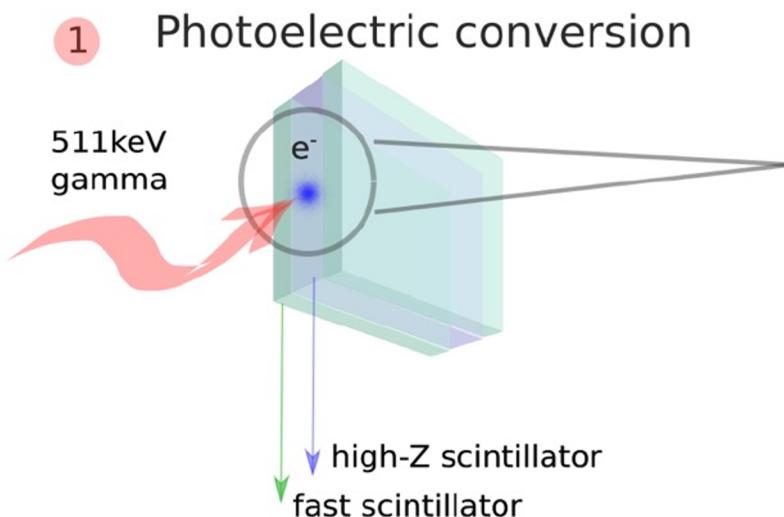


Toward detector designs

- Several developments are on going with nanocrystal scintillators to improve timing emission and light efficiency
 - How to exploit them for radiation detectors ?
 - Replace “classical” liquid scintillators by quantum dot in solution
 - Replace “classical” plastic scintillators by quantum dots embedded in Polymer/Glasses
 - Deposit quantum dot/nanoplatelet to a passif substrate or active material
- ⇒ heterostructure material: combine various materials with various properties
- ⇒ Such as layer bulk & nanomaterial scintillator or embedded in host material
- ⇒ Play with various size quantum dots to have a multispectral response
- ⇒ Exploit development of new material shape available:
- ⇒ 3D printing
- ⇒ micropulling down technique to grow crystal fibre or crystal tubes

Heterostructure concept

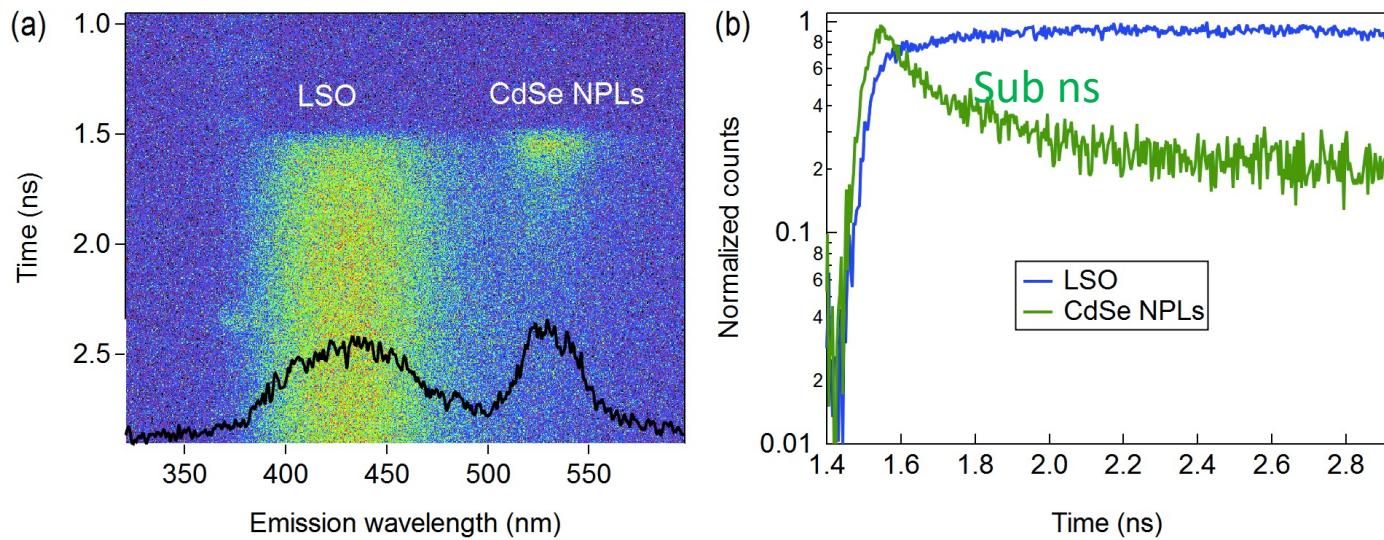
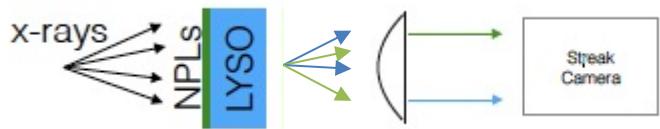
Combine scintillators with high light yield, high stopping power & material with prompt emission



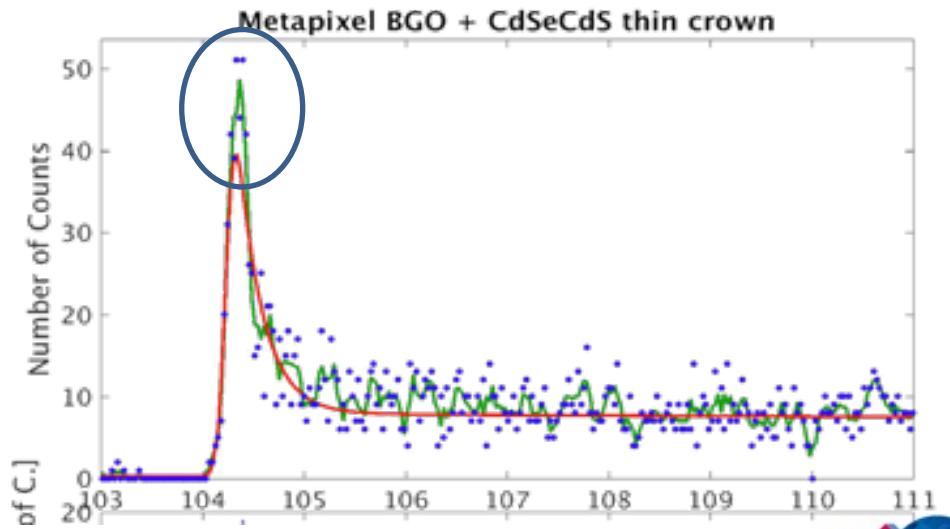
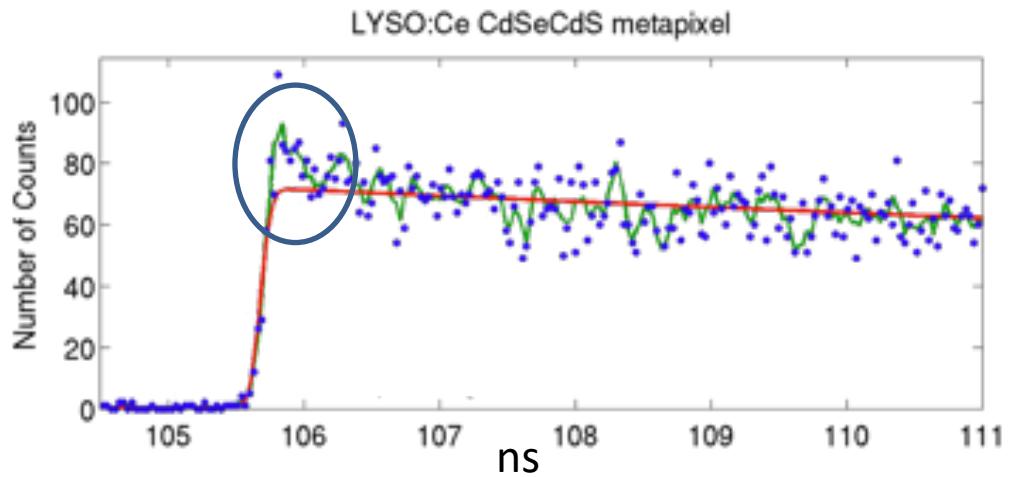
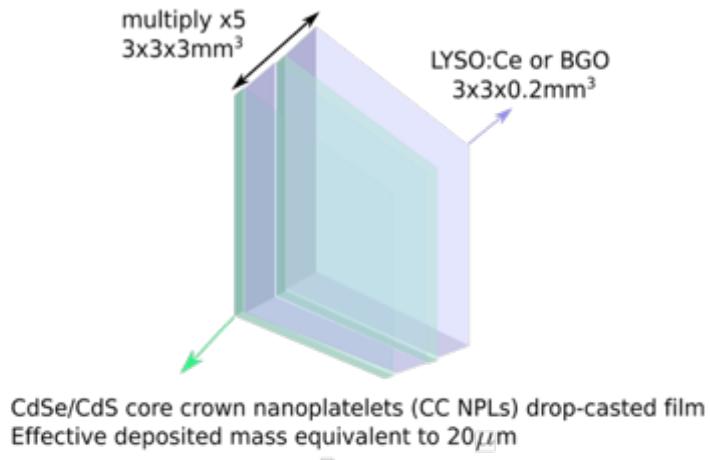
For some events, the energy of the recoil e- can be deposited in both materials

Energy sharing among multiple layers of standard and fast scintillators

CdSe nanoplatelets deposited on LYSO

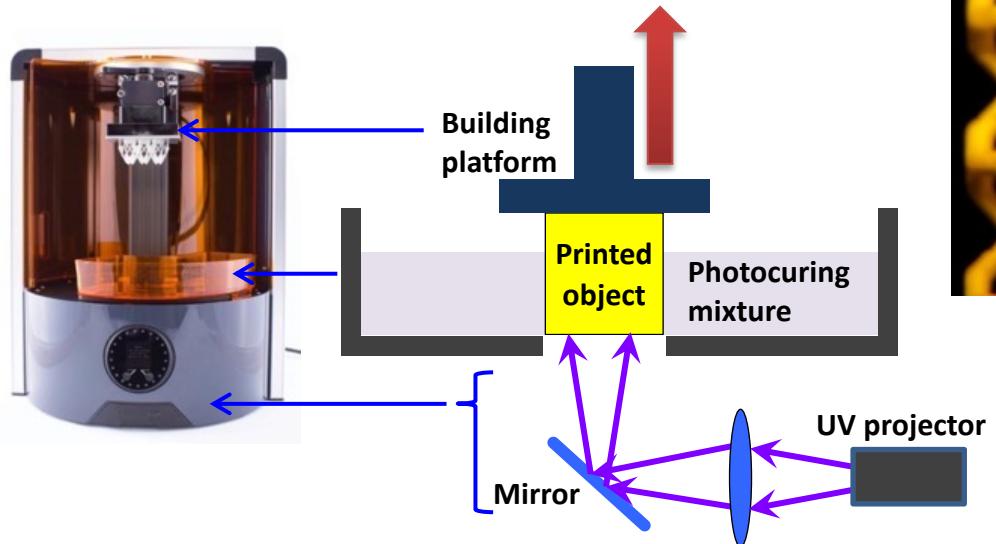


First attempt of heterostructure realized in our group

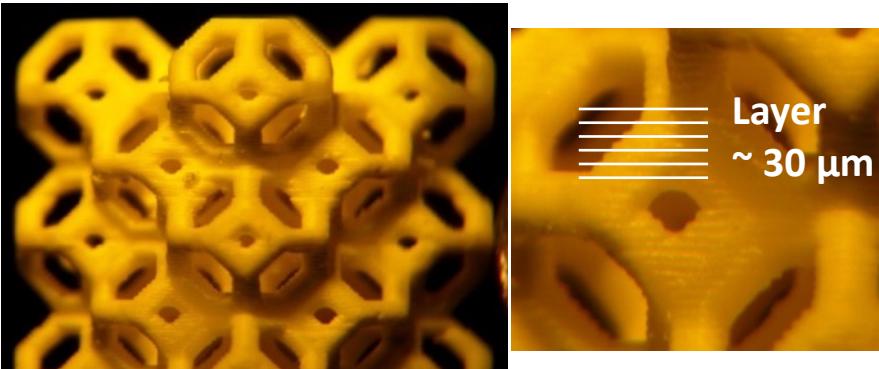


New production method: 3D printing

A way to design detector with unconventional shape



Printing is done layer-by-layer
Voxel size is $\sim 50 \times 50 \times 10-50 \mu\text{m}$

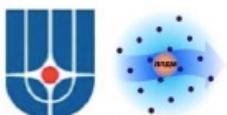


YAG



YAGG

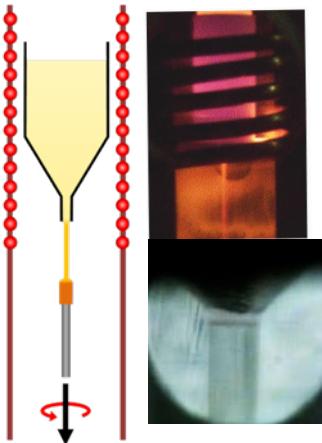
Hole $\varnothing < 400 \mu\text{m}$



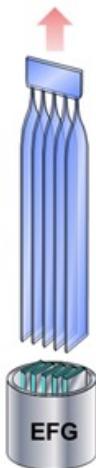
Courtesy of G. Dossovitsky, Kurchatov Institute

Crystal fiber production

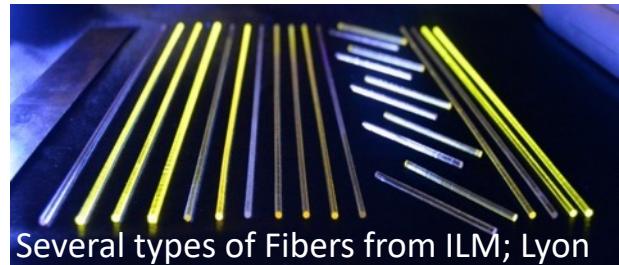
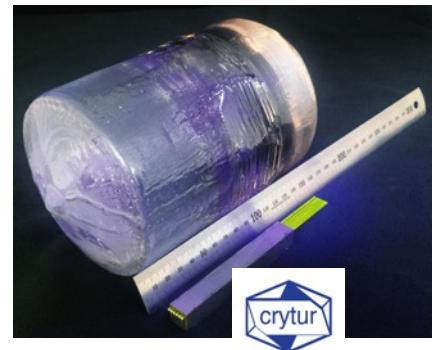
Micropulling down technique



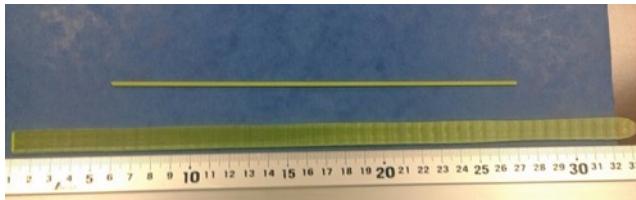
EFG



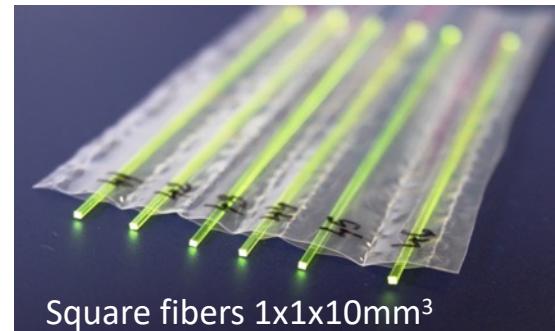
Czochralski method
Cut from large ingot



Several types of Fibers from ILM; Lyon



EFG-grown plate & fiber of LuAG:Ce
from Adamant Namiki Co , Japan



Square fibers 1x1x10mm³

⇒ Feasibility study: in an ANR project INFINI (ILM Lyon, CERN) and Intelum project (European Rise grant 644260) with 16 Partners (many from CCC) from 12 different countries: 11 academia and 5 companies



A lot of R&D is needed

To understand

- The scintillation mechanism in nanomaterial as we had to do 30 years ago on bulk scintillators
 - Excitation process and emission process
 - Role of the size of nanoparticles
 - Energy transfer from host to nanoparticles in case of embedded material
 -
- How to reduce self absorption
- How to improve
 - stopping power
 - Light transport
 - Chemical stability
- How to build/optimize Heterostructure material: eg combined organic and inorganic components
- Development of large production scale
 - Some developments of large scintillating materials based on quantum dots already exist (eg solar concentrator)

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