Fast Advanced Scintillator Timing
(2014-2018)

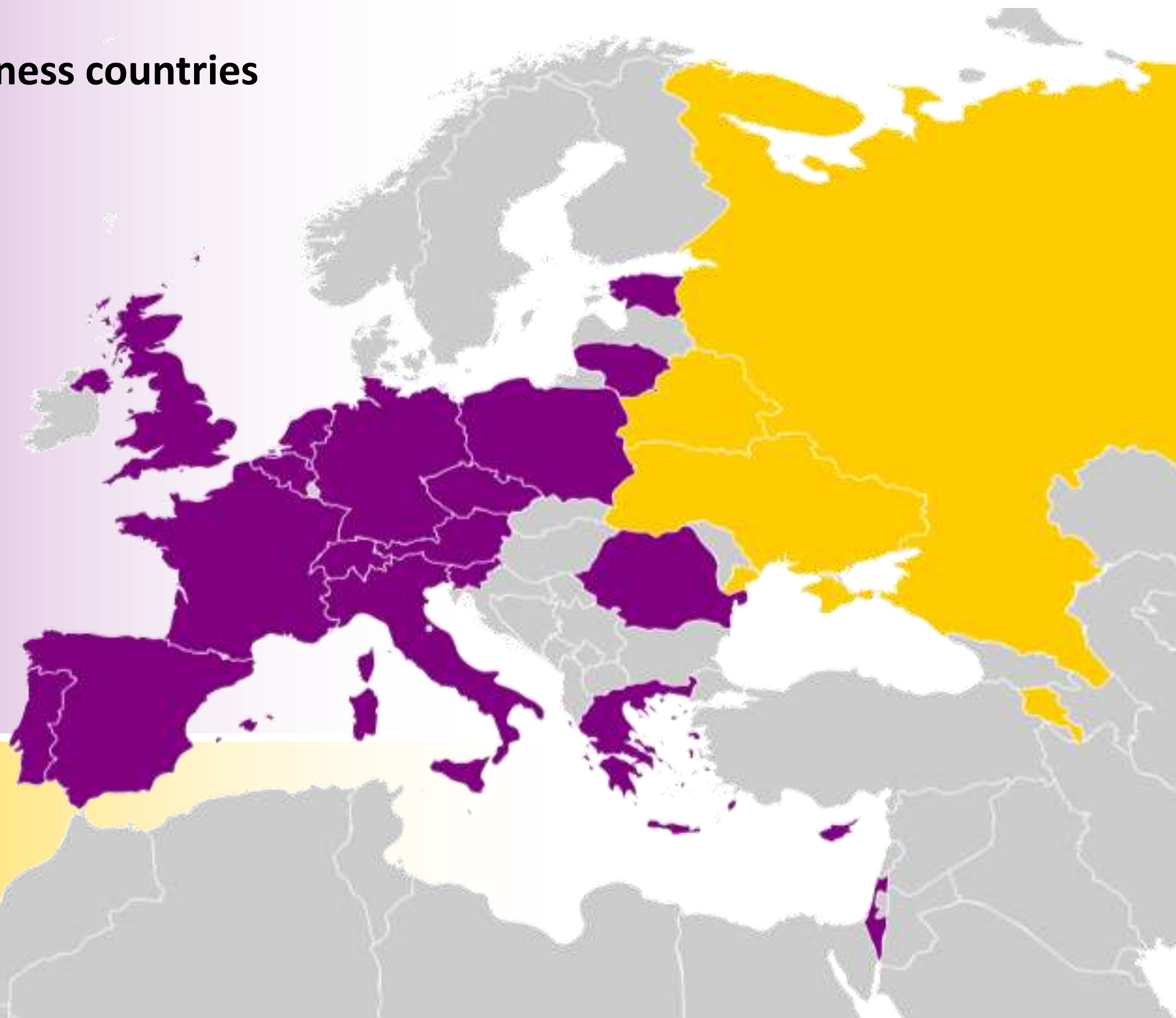
COST Action TD1401: Nanocrystalline and nanocomposite scintillators for fast timing

M. Nikl on behalf of COST FAST, TD1401 project consortium
Institute of Physics, AS CR, Prague, Czech Republic

FAST Today participating countries

(19 COST and 4 Near Neighbours)

8 inclusiveness countries



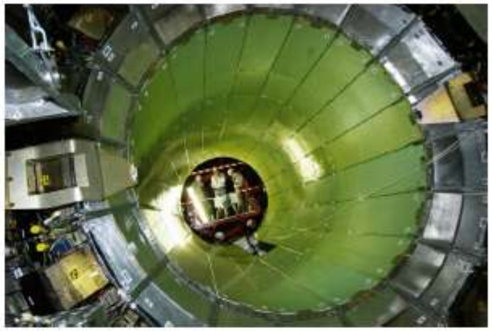
COST countries

- Austria
- Belgium
- Cyprus
- Czech Republic
- Estonia
- France
- Germany
- Greece
- Israel
- Italy
- Lithuania
- Netherlands
- Poland
- Portugal
- Romania
- Slovenia
- Spain
- Switzerland
- United Kingdom

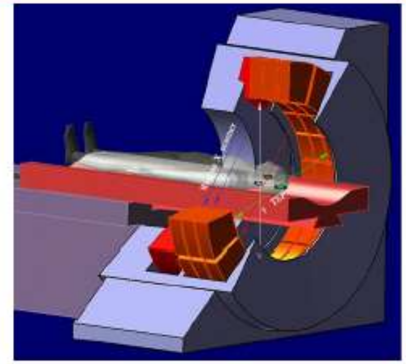
COST Near Neighbour Countries

- Armenia (1 institute)
- Belarus (1 institute)
- Ukraine (1 institute)
- Russian Federation (3 institutes)

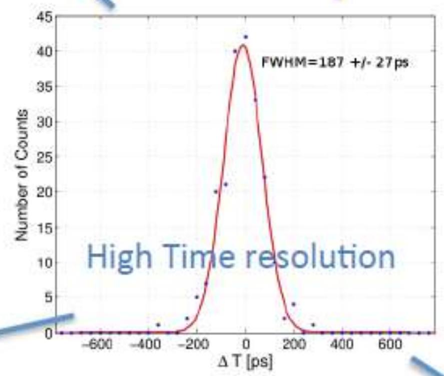
Areas benefitting from high time resolution



High Energy Physics Calorimetry



Positron Emission Tomography



High Time resolution

Quantum Cryptography

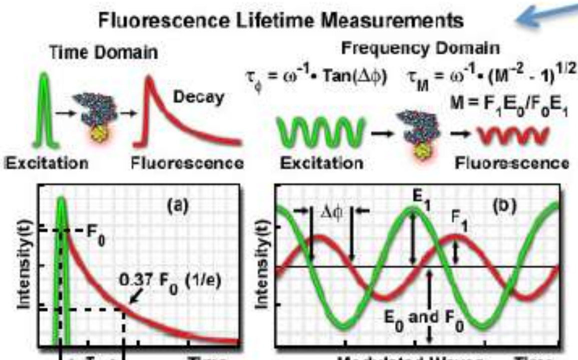
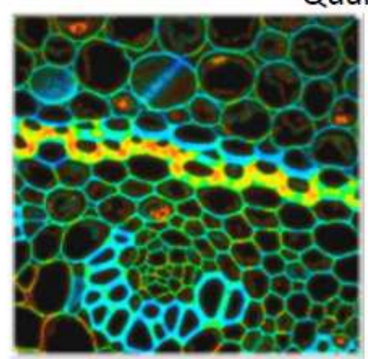


Figure 10

FLIM: Fluorescence Lifetime Imaging Microscopy

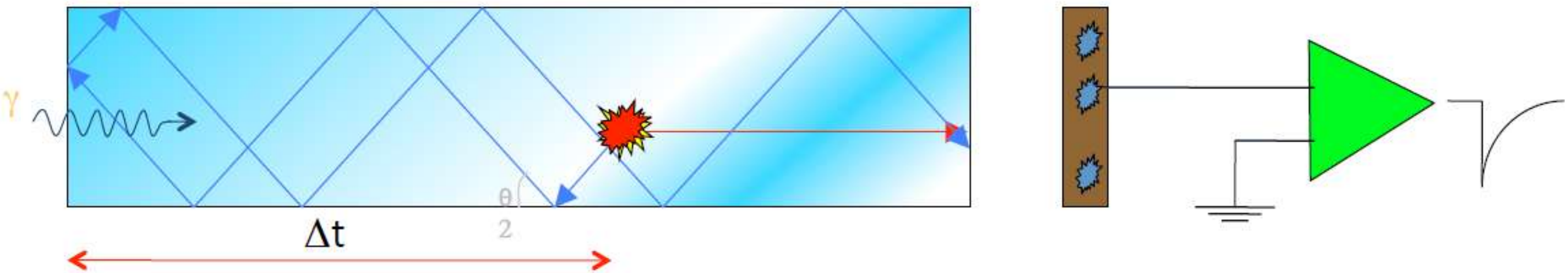
&

FRET: Förster Resonance Energy Transfer



The Photodetection Chain

Crystal | **Photodetector** | **Electronics**



$t_{kth\ pe} = \Delta t + t_{k'\ ph} + t_{transit} + t_{SPTR} + t_{TDC}$

- Δt: Conversion depth
- $t_{k'\ ph}$: Scintillation process
- $t_{transit}$: Transit time jitter
- t_{SPTR}: Single photon time spread
- t_{TDC}: TDC conversion time

TODAY

WG 2

Scintillator R & D

- Interaction
- Light generation
- Light transport
- Light transfer
- Light collection

WG 3

Photodetector R & D

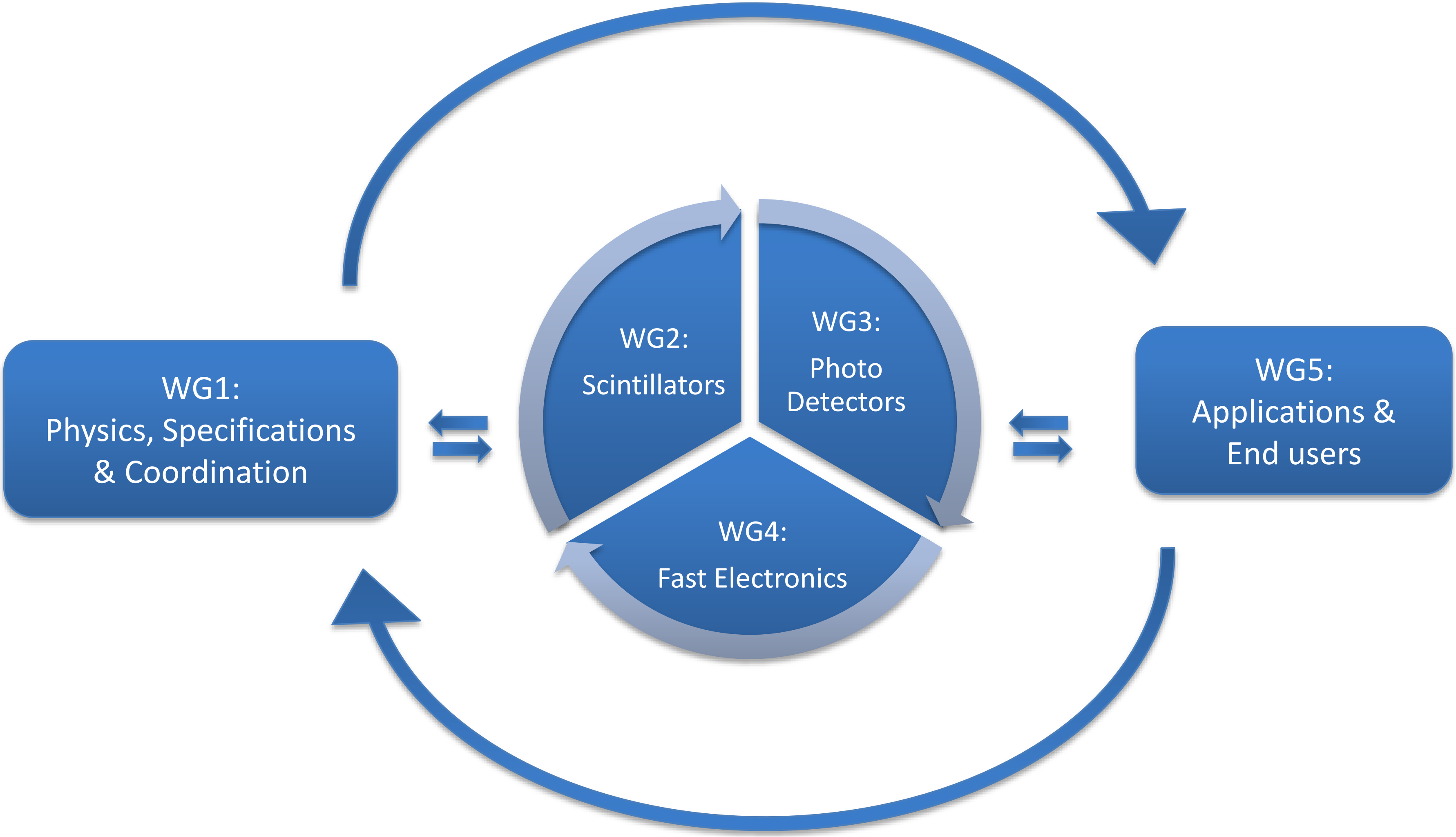
- Reduce SPTR and DCR
- Increase fill factor (PDE)
- Digital SiPM
- MCP for PET & HEP

WG 4

Electronics R & D

- TDC < 10ps bins
- Monolithic architecture
- High bandwidth
- Low noise
- Massive parallel data
- High number of channels

Working groups interaction



Exchanges through meetings, STSM, workshops, projects

WG2: Critical parameters for fast timing:

- ❑ A high light yield
- ❑ Short decay time
- ❑ As short as possible rise time
- ❑ Light transport in the sample before reaching photodetector

Squeezing photons to arrive to photodetector in the **as short as possible** time window !!!

Due to stochastic characteric of processes in the initial relaxation (conversion stage), transport of charge carriers (transfer stage) and lifetimes of emission centers (typ. above 10 ns) it is difficult in classical (Ce-doped or intrinsic wide band-gap materials (BGO, CeF3) achieve better time resolution then 100 ps

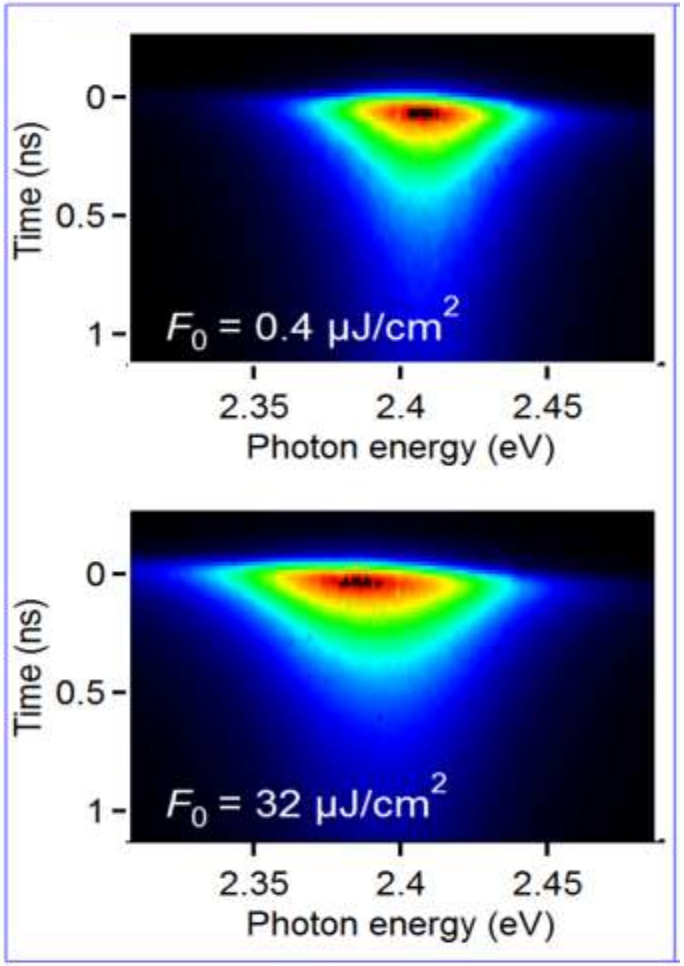
Types of emission in scintillating crystals and delay between energy deposit and photon emission

- Excitonic emission (STE, excitations of anion complexes)
- Emission of activators (Ce, Pr, …)
- Crossluminescence
- Quantum confinement driven luminescence
- Intraband hot luminescence
- Cherenkov radiation

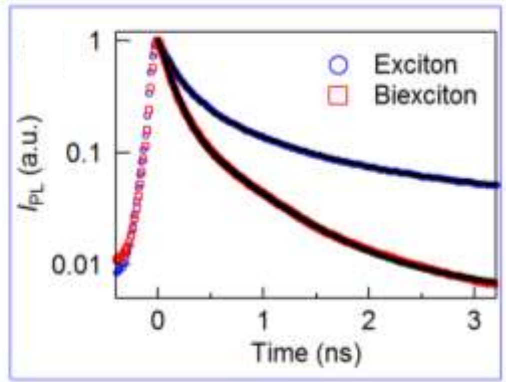
Slow

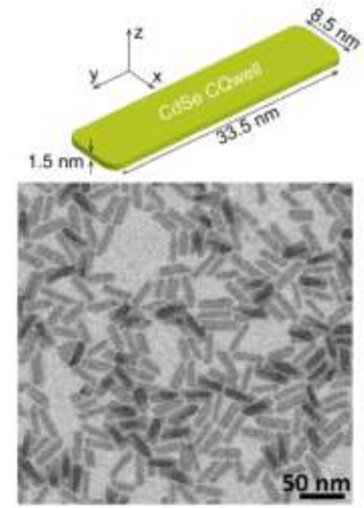
Short

Colloidal CdSe nanosheets (quantum wells)



Emission 500-520 nm
Exciton lifetime: **440 ps**
Biexciton lifetime: **125 ps**





- 1D Quantum confinement implies strict selection rules
- In-plane delocalization implies fast exciton recombination rate (giant oscillator strength transition).
- Strongly suppressed Auger recombination in 2D CQwells.

Increased exciton coherence volume ⟶ Reduced Auger $\tau_R^{-1} > \tau_{Auger}^{-1}$

↓

Enhanced emission rate: $\tau_R^{2D} \propto \frac{\Delta\lambda}{E_b^{2D}}$

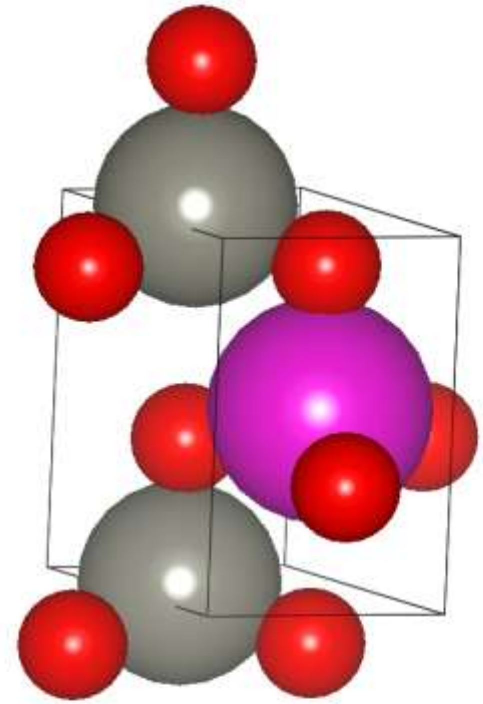
J. Grim et al. *Nature nanotech.* **9**, 891–895 (2014)

ZnO-based nanocrystals

- Hexagonal structure of wurtzite
- Usually non-stoichiometric $Zn_{1+x}O$; n-type semiconductor – naturally doped by O vacancies and Zn interstitials
- Advantageous properties–**high radiation stability**, absorbance in UV and transparency in visible spectral range
- Optoelectronic properties– wide band gap (3,4 eV), **high E_B of excitons (60 meV)**, low afterglow, **extremely short luminescence decay of excitons (sub-ns)**

Possible applicability: optical fibers, photovoltaic devices, **high-energy radiation detectors**…

Solid solutions $Zn_{1-x}Cd_xO$, $Zn_{1-x}Mg_xO$



- Possibility of Cd^{2+}, Mg^{2+} incorporation into the ZnO structure; substitution of Zn^{2+} ions

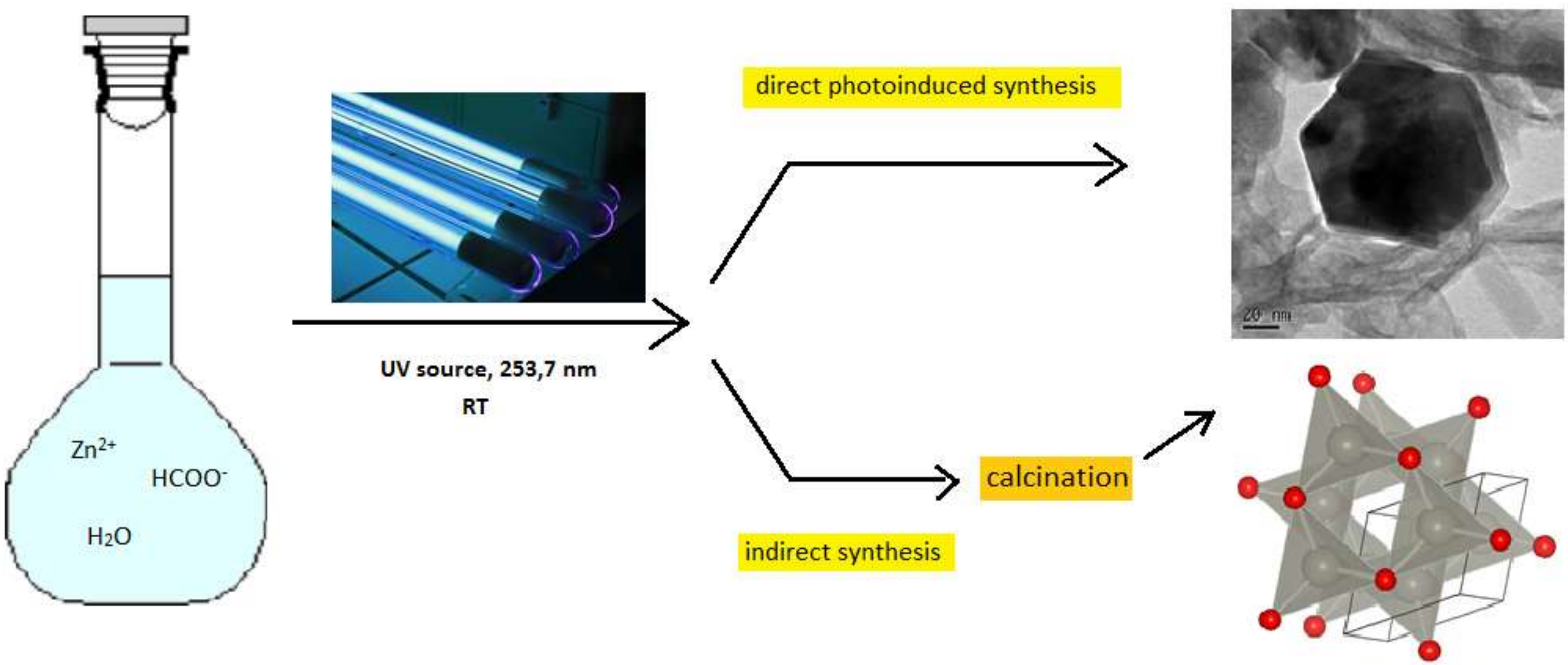
  Crystal radii:
  Zn^{2+} (coord.num.IV): CR = **0,74 Ǻ**
  Cd^{2+} (coord.num.IV): CR = 0,92 Ǻ
  Mg^{2+} (coord.num.IV): CR = 0,71 Ǻ

- Solubility limit of Cd^{2+} in the ZnO structure is 2%, but SS with Cd^{2+} and Mg^{2+} content up to 25% resp. 30% Mg is possible (Makino et al., 2001, Lange et al., 2012)
- Cd/Mg content depends on the preparative technique

Photo-induced synthesis-overview

Radiation- or photo-induced precipitation:

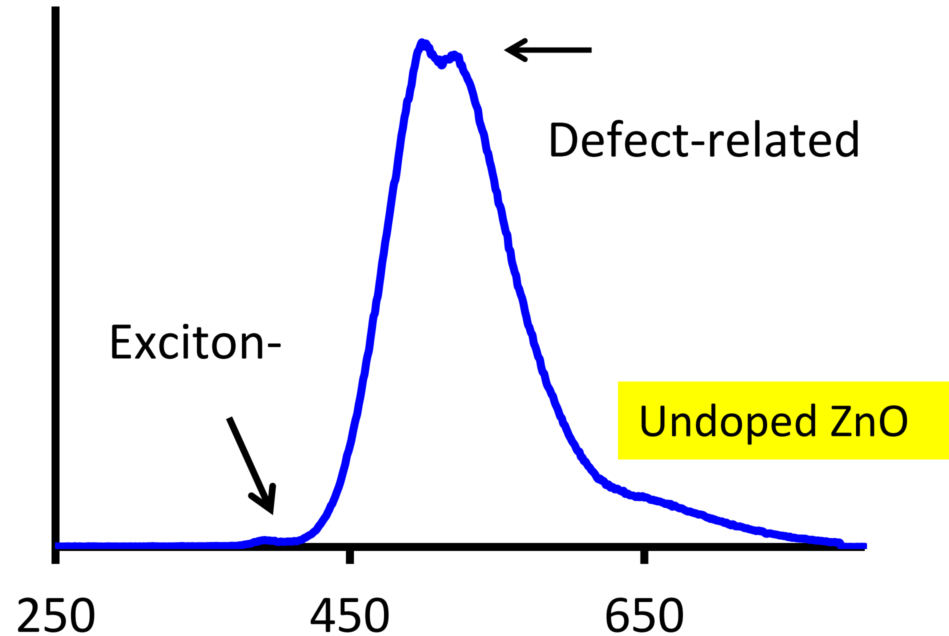
Principle: reaction of dissolved precursors with products of radio/photolysis of water leading to the precipitation of solid phase (particle size~nm)



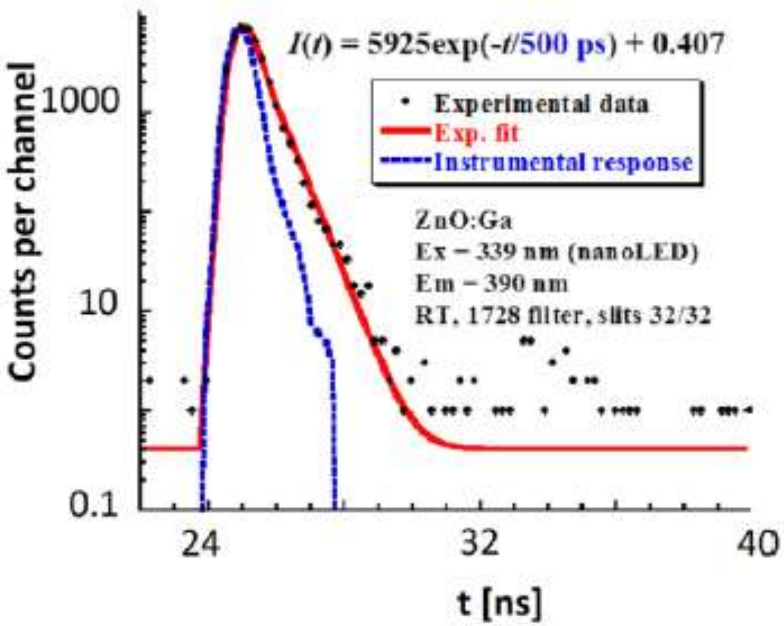
Prepared by V. Cuba et al, CTU Prague

Luminescence characteristics of powder

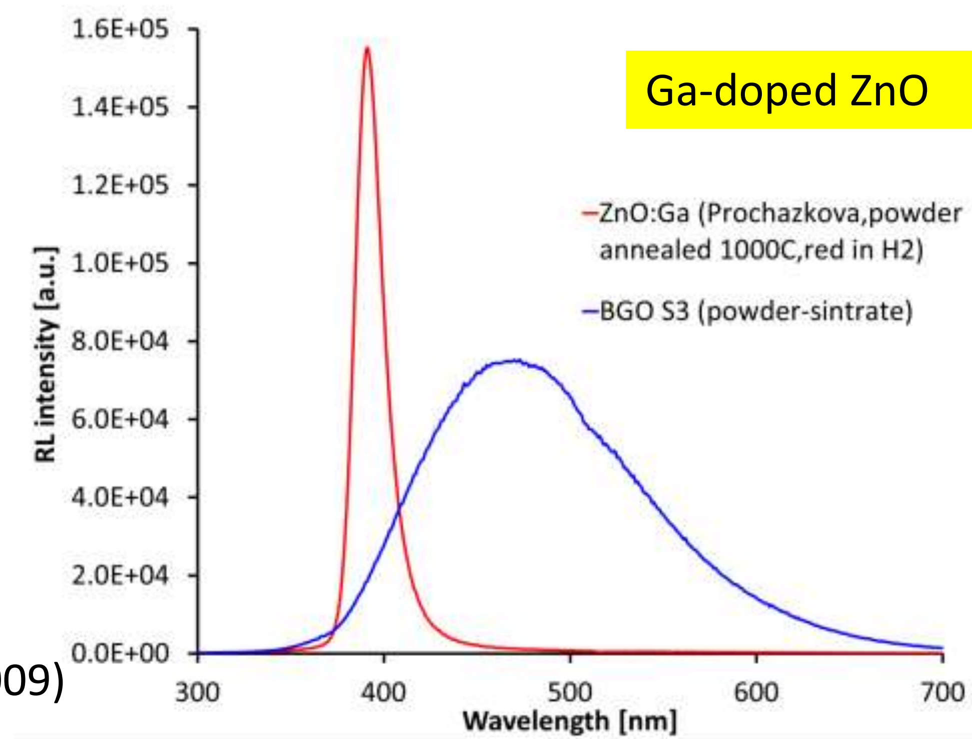
Subnanosecond decay of exciton state is a suitable center for superfast scintillator!



PL decay of Ga-doped ZnO exciton emission

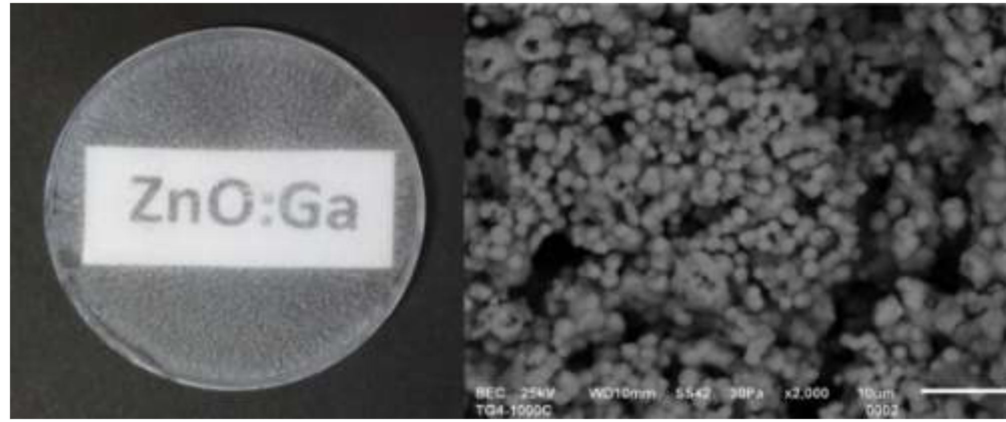


Bourret-Courchesne et al, NIM A 601, 358 (2009)



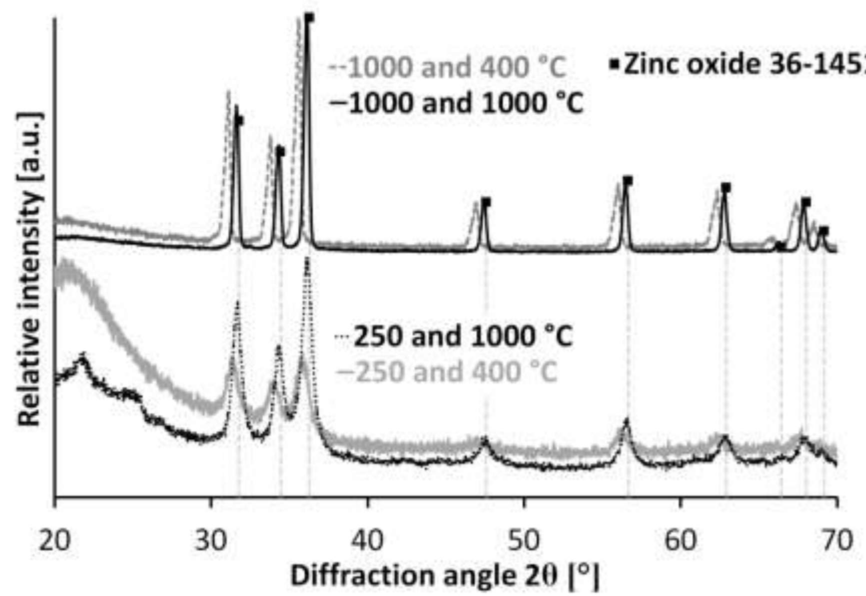
Composite materials I.

ZnO:Ga-SiO_2 composite on glass substrate



- **Effect on annealing temperatue**:
  a) powder ZnO:Ga during its preparation – 250 C and 1000 C;
  b) Zno:Ga embedded into the SiO_2 matrix – 400 – 1000 C => structural and luminescence properties?

- **Effect on structural properties**:
  optimal conditions were found, when there is no unfavorable interaction between ZnO and the matrix

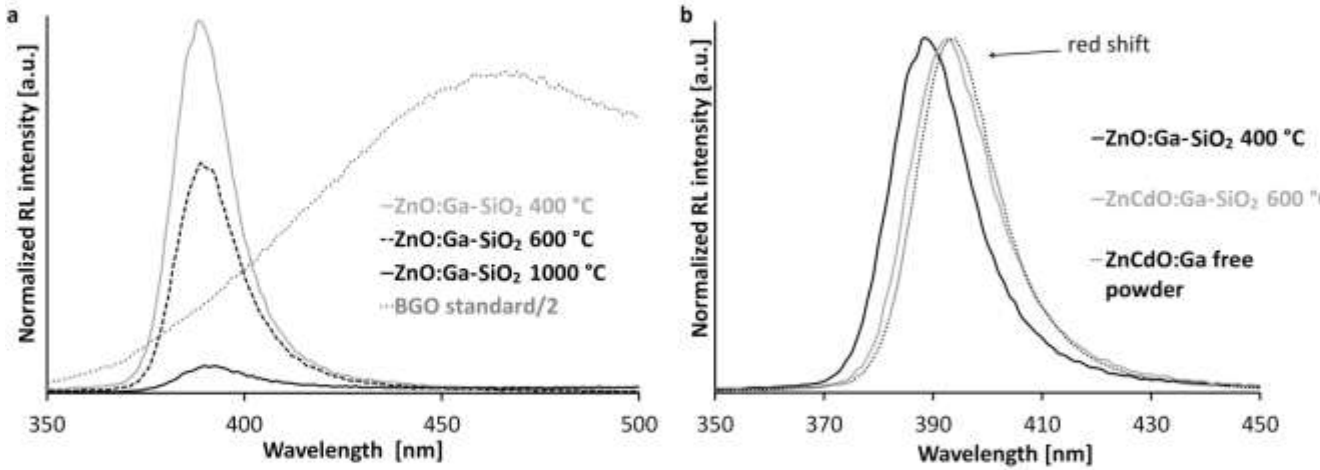


(Procházková et al., Preparation of Zn(Cd)O:Ga-SiO_2 composite scintillating materials – Rad. Meas. 2016)

Composite materials I.

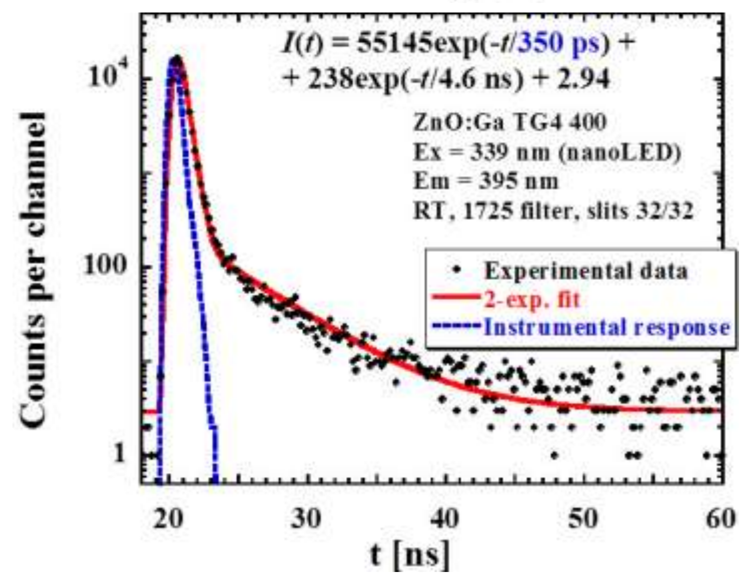
Effect on the luminescence properties:

- Decrease in RL intensity with the increasing annealing temperature



- **$Zn_{0.97}Cd_{0.03}O$:Ga-SiO_2** red shift of the excitonic-related emission, probably effect of band gap modulation

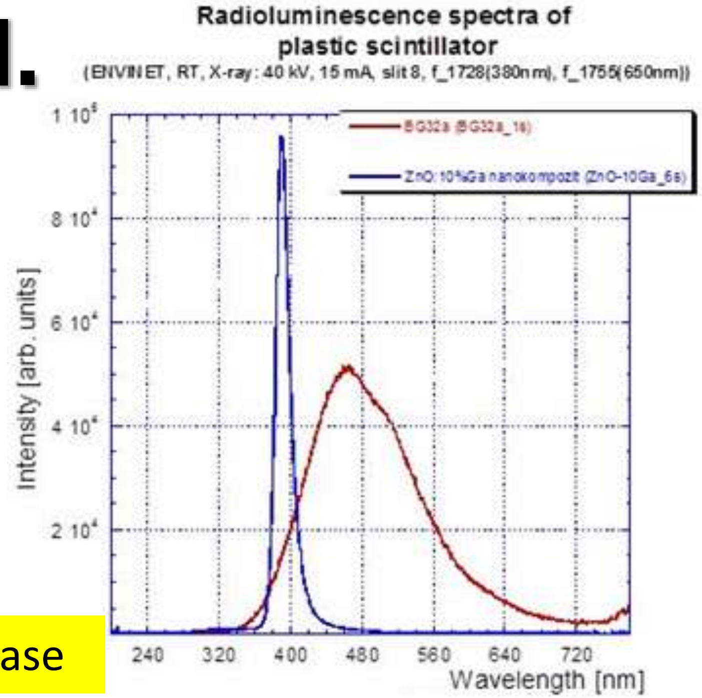
- **PL decay** – two components, ultrafast ~ 350 ps, „slow“ ~4,6 ns (capture on the deffects at ZnO:Ga – SiO_2 inteface (less than 10 %)



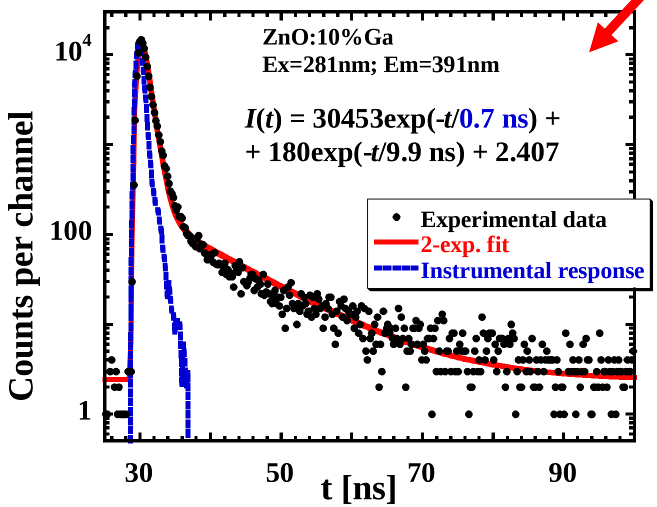
Composite materials II.

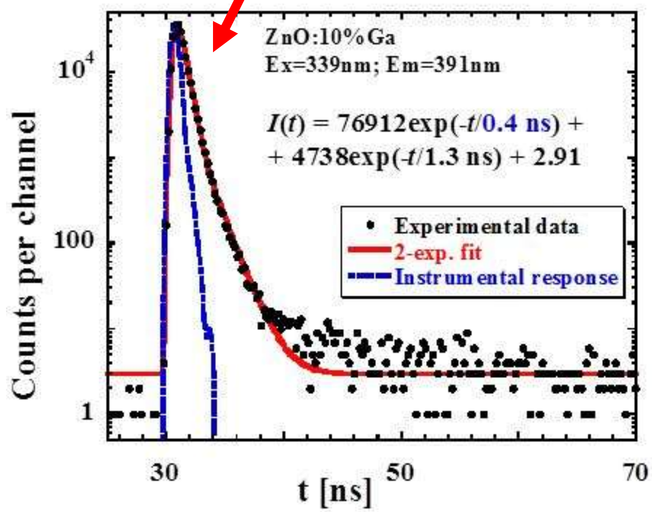
ZnO:Ga-PS (polystyrene matrix)

- 10 wt. % ZnO:Ga in PS matrix
- RL spectra – only ZnO:Ga emission
- PL decay - excited at 281 and 339 nm; nonradiative energy transfer ZnO:Ga – PS (~400 ps)

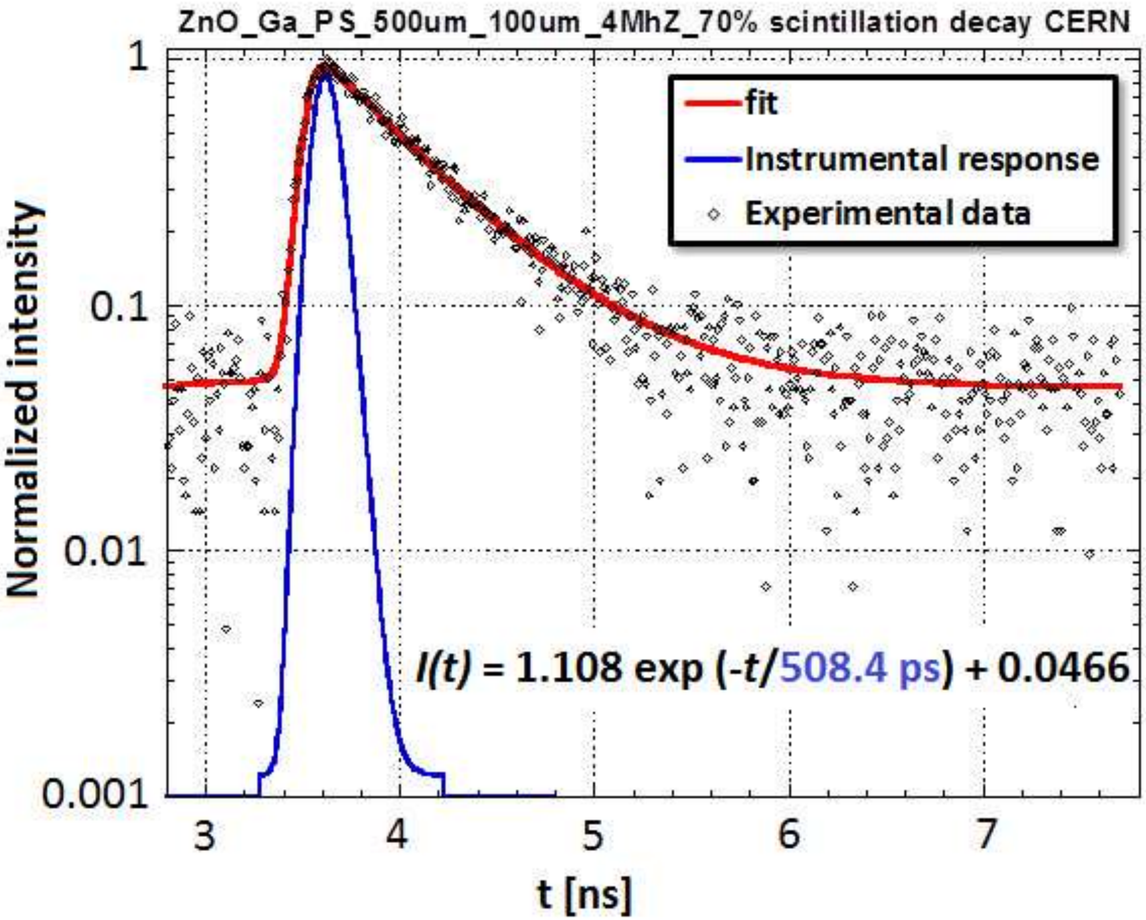


PL decays under excitation into PS host and ZnO phase





ps X-ray excited decay ZnO:Ga@PS composite



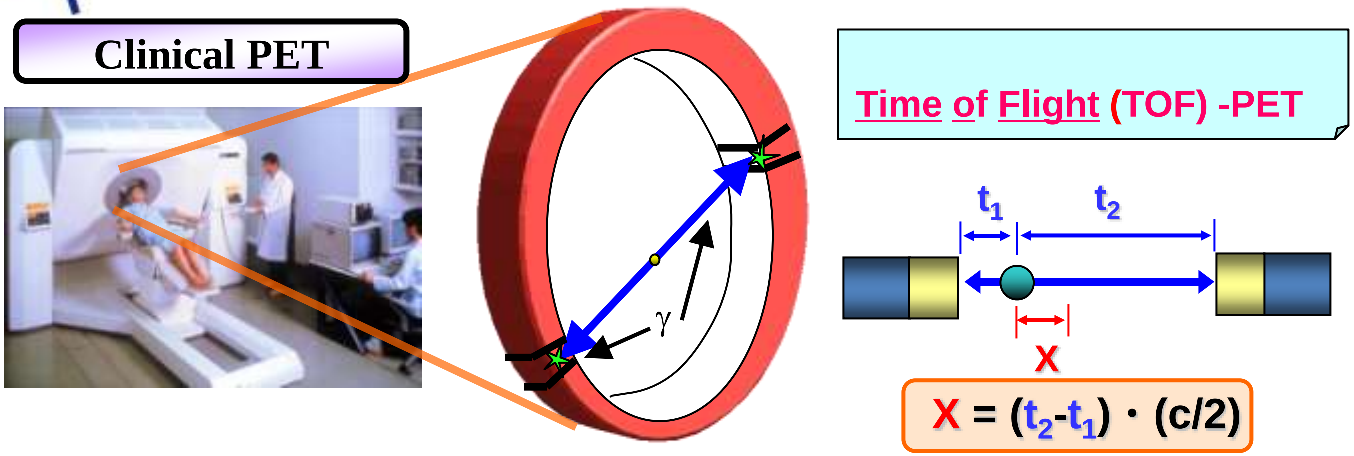
. Image of ZnO:Ga-PS composite 1 mm thick with 10 wt% of ZnO:Ga filling

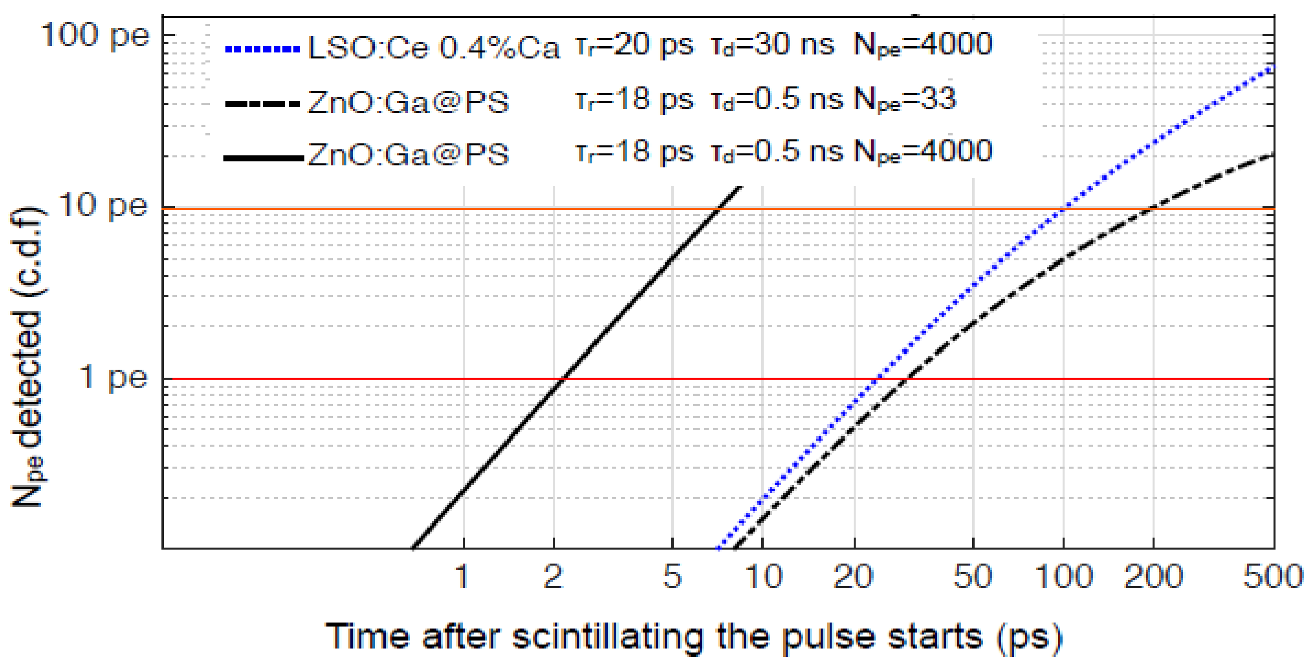
Rise time below the time resolution of the set-up (18 ps) !!!

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

TCR of ZnO:Ga@PS composite

For time-of-flight techniques “timing coincidence resolution” (TCR) is important





Using 22Na source the comparison of ZnO:Ga@PS versus Lu2SiO5:Ce 0.4% Ca co-doped is done by calculating the photoelectron cumulative distribution functions along the first 500 ps of the scintillating pulse.

Turtos et al, submitted to phys.stat.sol. RRL

Low Npe of ZnO:Ga@PS is mostly due to low transparency and low QE

An analogous situation exists in 1D-confined multiple quantum well (MQW) nanostructures

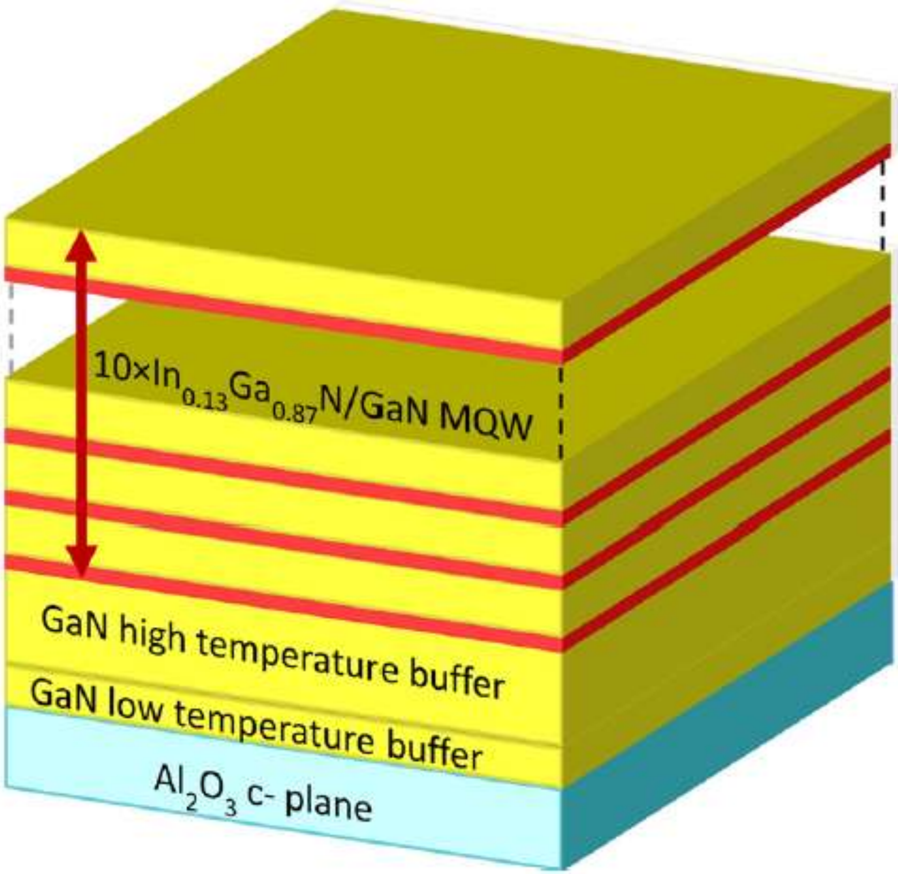
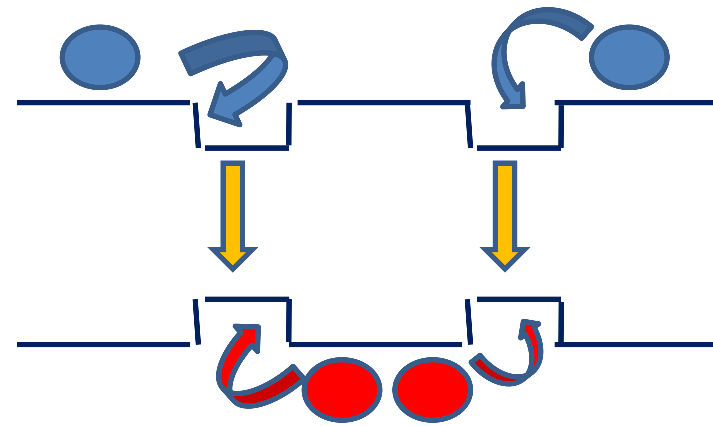


Figure 1. A schematic drawing of the multiple quantum well structure.

Hospodkova et al, Nanotechnology ***25****, 455501 (2014).*

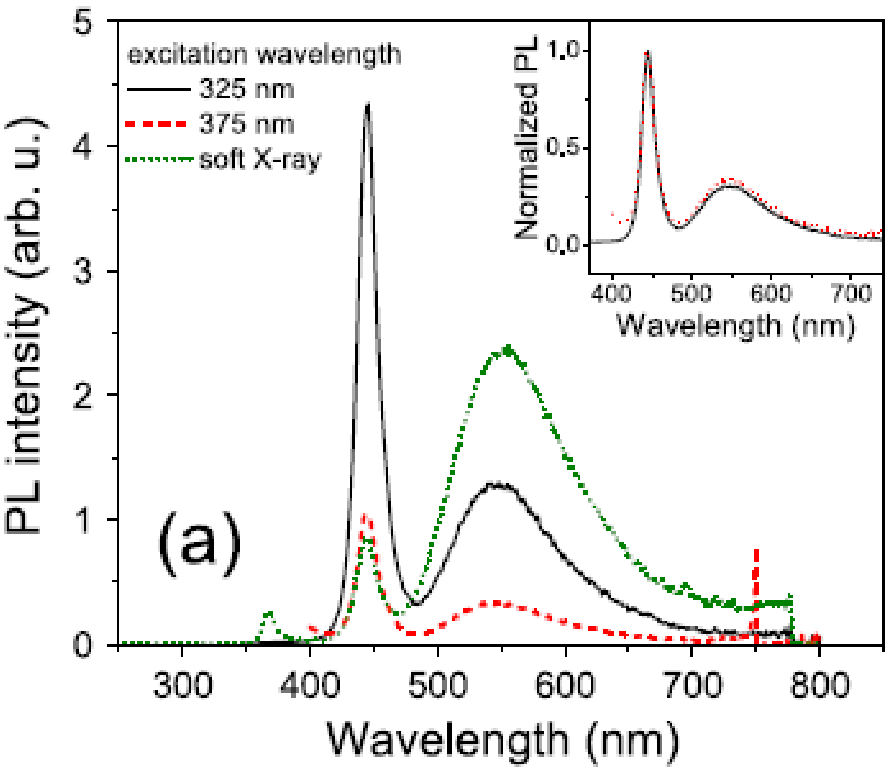


Electrons and holes are concentrated in narrow gap layers and radiatively recombine there being spatially confined by small thickness (few nm) of the layer

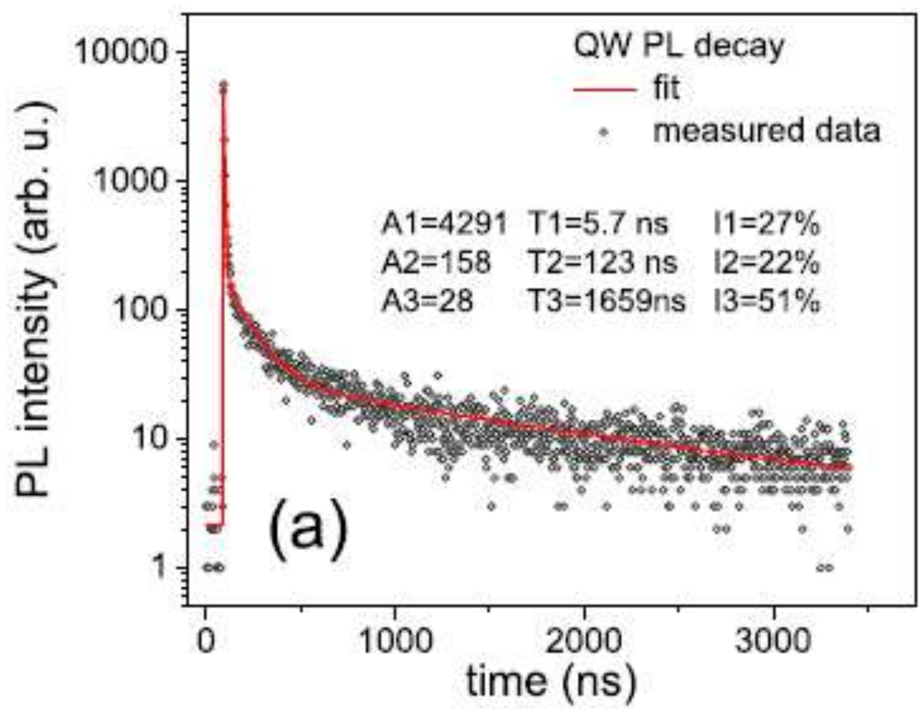
MOVPE technology can prepare such nanostructures on 4-6 inch size Al2O3 substrates

Spectral characteristics of GaN-GaInN MQW

Luminescence spectra



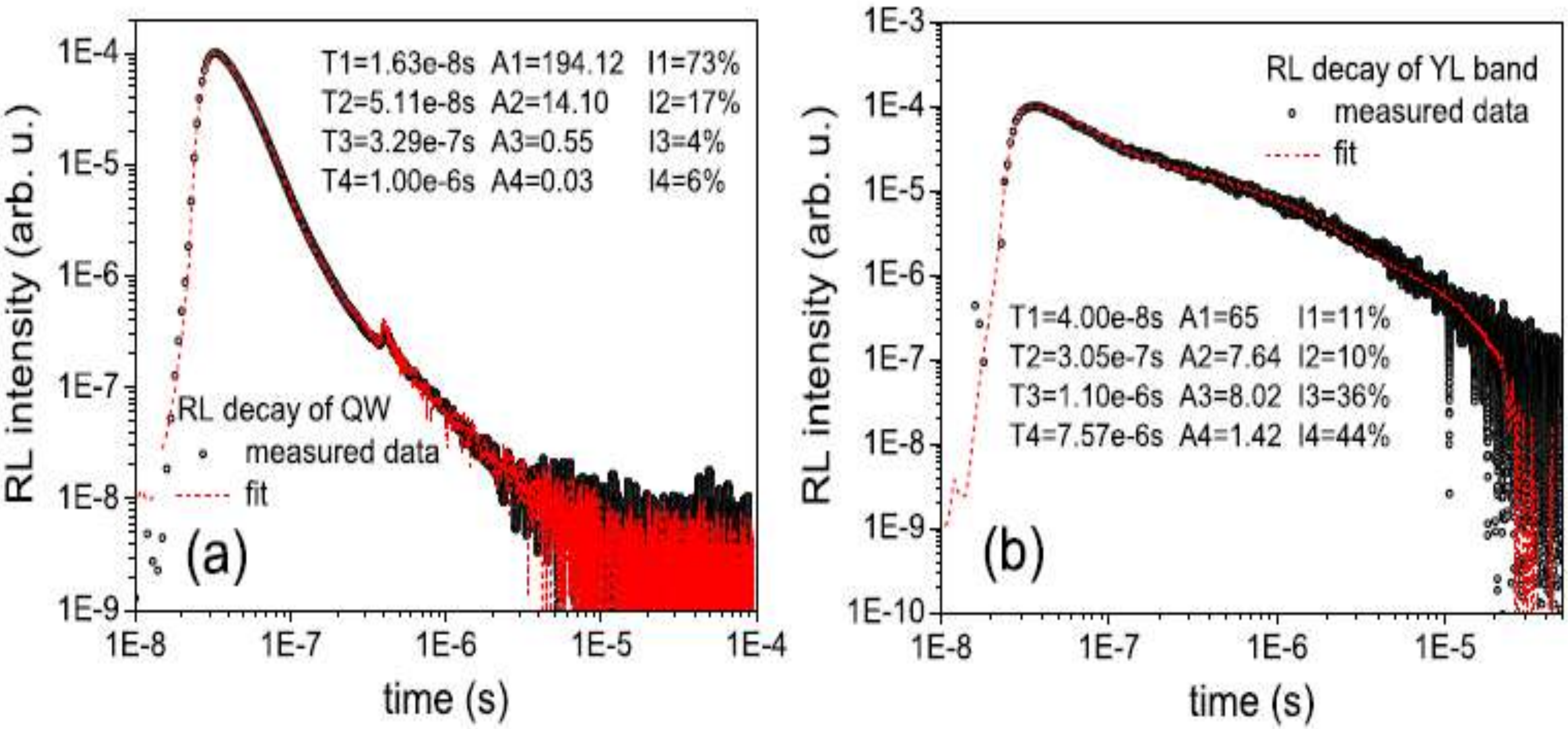
Photoluminescence decay



The problem is the defect-based emission band in yellow spectral region. Optimization of MQW shape and composition can bring PL decay time down to about 1 ns preserving high QE.

Scintillation response of GaN-GaInN MQW

Scintillation decay excited by ns pulse of soft X-ray (300-400 eV)



While the dominant component in exciton band is about 16 ns, in the yellow band it spans down to microsecond time scale!

Intraband luminescence (IBL) mechanism

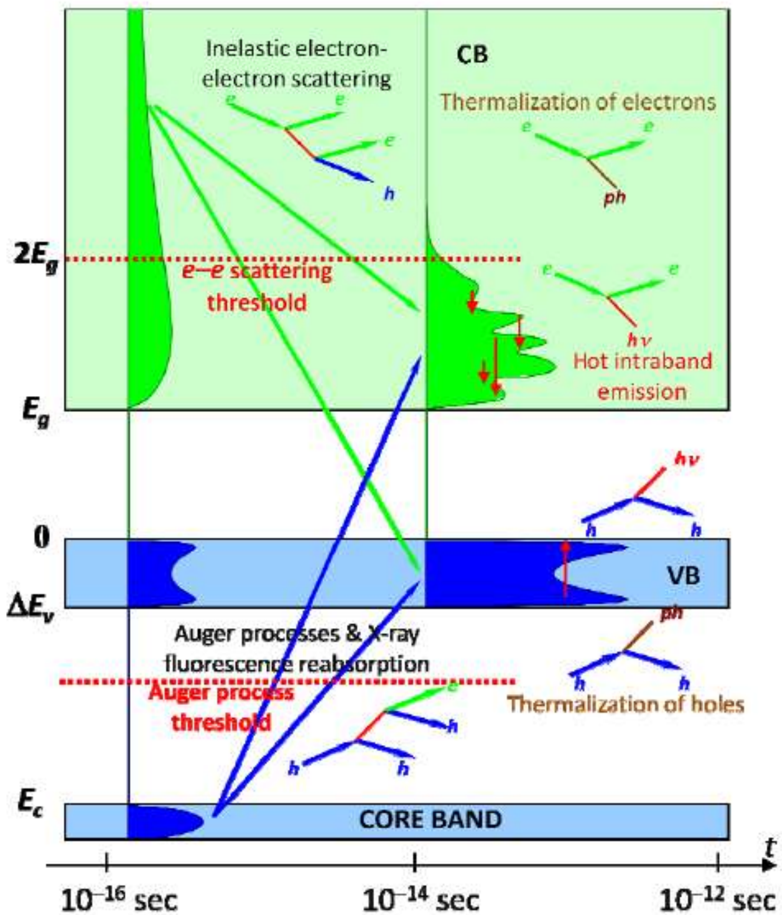


Figure: The mechanism of IBL (drawing by A. Vasil'ev)

- Decay 10^{-12} s
- Electron component (e-IBL): broad spectrum through all transparency range
- Hole component (h-IBL): spectrum covers VB width

Experimental NIR spectrum of IBL

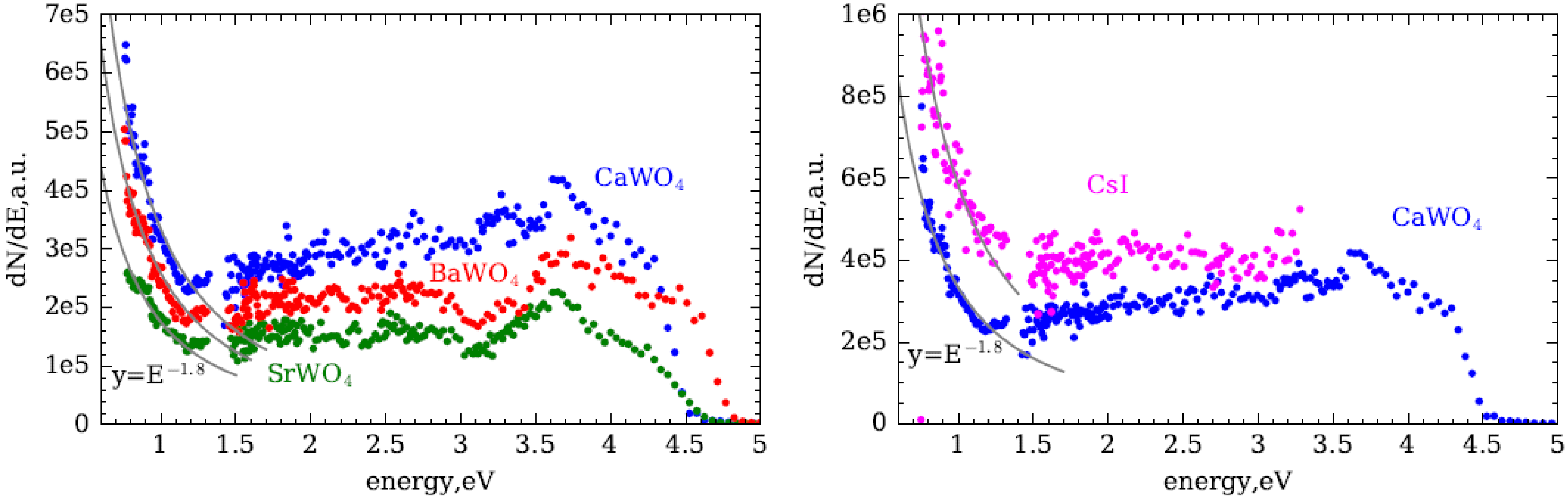


Figure: Spectral distribution of the number of prompt photons emitted by sample during electron pulse. Normalized arbitrarily.

Light yield of IBL

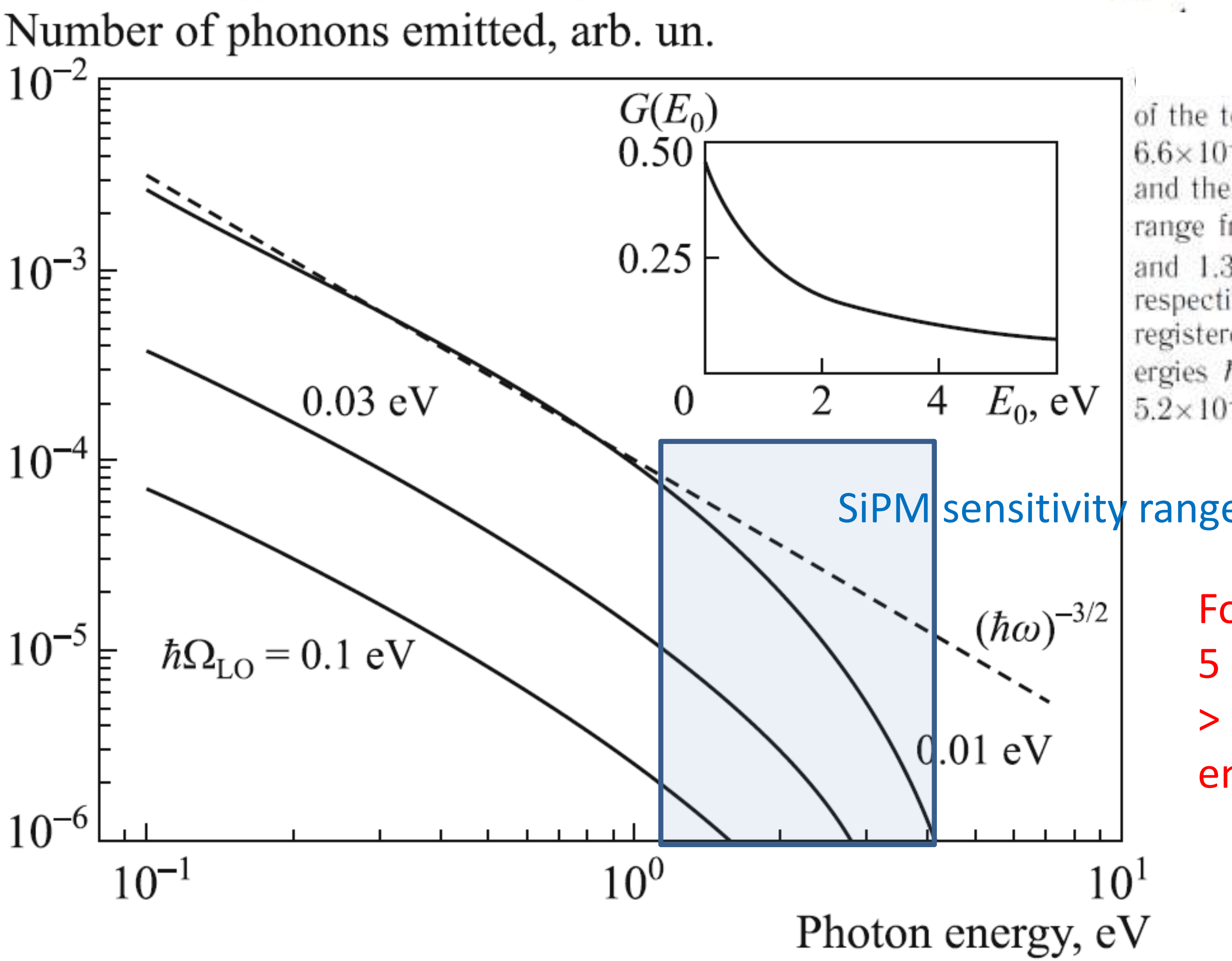
Compound	LY@662keV ph/MeV	LY@50keV ph/MeV	experi-mental LY ph/MeV	IBL LY >1.5 eV
LYSO	33000	26400 (0.8)	=26400	<19
BGO	8200	8200	9520	19
CeF_3	4500	3375 (0.75)	3022	12 (<4 eV)
PWO	100–200	-	160	18

LY values measured at these materials are too low …

A.N.Vasil'ev, R.V.Kirkin, Physics of Wave Phenomena, 2015, 23, 186-191

Emission spectrum of intraband luminescence for single parabolic band under excitation of wide-band-gap insulators by ionizing radiation and particles

$$n(\hbar\omega) = 1.5,\ m_e = 0.5m_0 \quad E_g = 6\ \mathrm{eV}$$



The values of the total energy lost in the radiative channel are 6.6×10^{-6}, 3.7×10^{-5}, and 2.7×10^{-4} eV per electron and the numbers of photons emitted in the spectral range from $\hbar\Omega_{LO}$ to E_g are 1.3×10^{-5}, 1.2×10^{-4}, and 1.3×10^{-3} for $\hbar\Omega_{LO} = 0.1$, 0.03, and 0.01 eV, respectively. The numbers of photons that can be registered with silicon photomultipliers (with energies $\hbar\omega > 1.1$ eV) are 1.4×10^{-6}, 7.5×10^{-6}, and 5.2×10^{-5}, respectively.

For CsI (0.01 eV phonon)
5 emitted photons with energy > 1.1 eV per MeV of absorbed energy

Conclusions

- Wannier exciton-based emission combined with quantum size effect can be used to create superfast nanoscintillators (decay time < 1 ns) under the assumption of efficient suppression of surface losses at nanocrystals
- Embedding such nanocrystals (quantum dots) into a suitable host, where efficient and (super)fast energy transfer host->nanocrystal is achieved and transparency is preserved, can open the way for their practical use in hybrid scintillators for fast timing
- 1-d confined MQW systems as GaN-GaInN have also application potential in this field, if the radiative electron-hole recombination in MQW structure can be tuned to subnanosecond time scale

This research is supported by COST Action (TD1401, FAST), supported by European Cooperation in Science and Technology