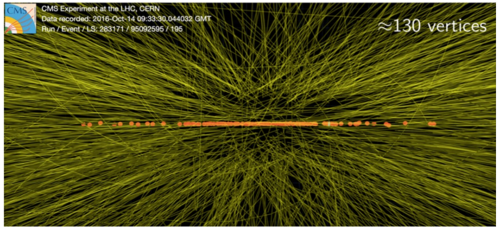
RECENT DEVELOPMENTS IN THE FIELD OF SCINTILLATORS FOR FAST RADIATION DETECTORS

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

Why FAST timing ?

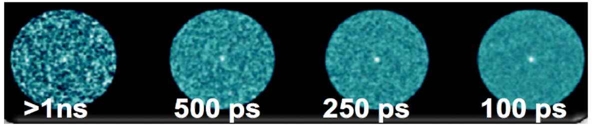
In HEP : Search for rare events implies high luminosity accelerators

→ Rate problems;
→ Pileup of >140 collision events per bunch crossing at *High Luminosity-LHC;*
→ Pileup mitigation via TOF requires TOF resolution < 50ps.



In Positron emission tomograph: Time of flight PET

→ Better image quality for same acquisition time
→ Faster exam
→ Simplify reconstruction
→ Help for limited field of view



=> Need to push the limit of time resolution of detectors

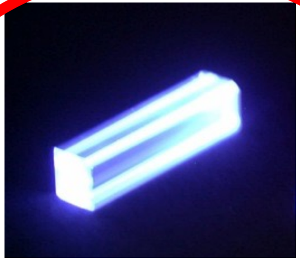
The full detection chain impacts timing properties

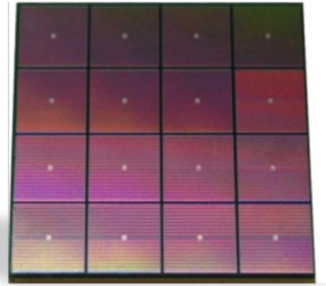
Light production & transport

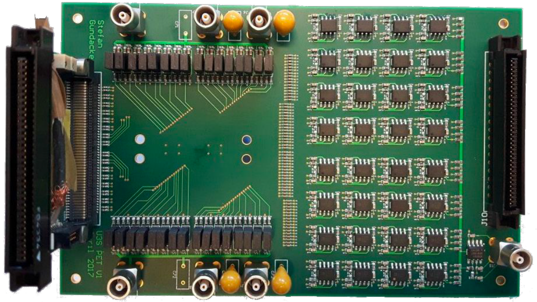
Light detection

Electronic readout

γ



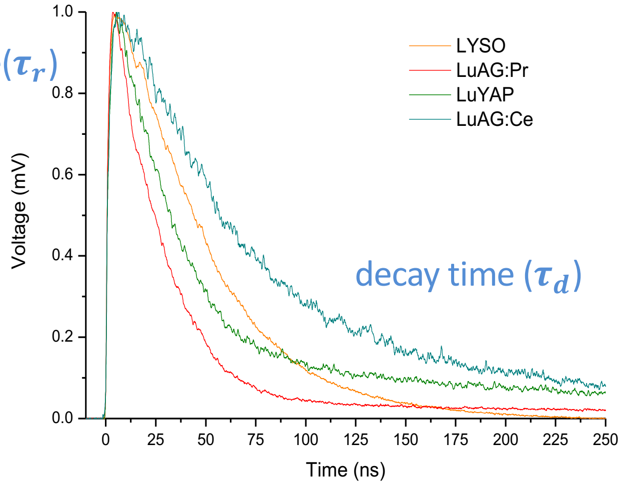




Many developments ongoing on scintillators

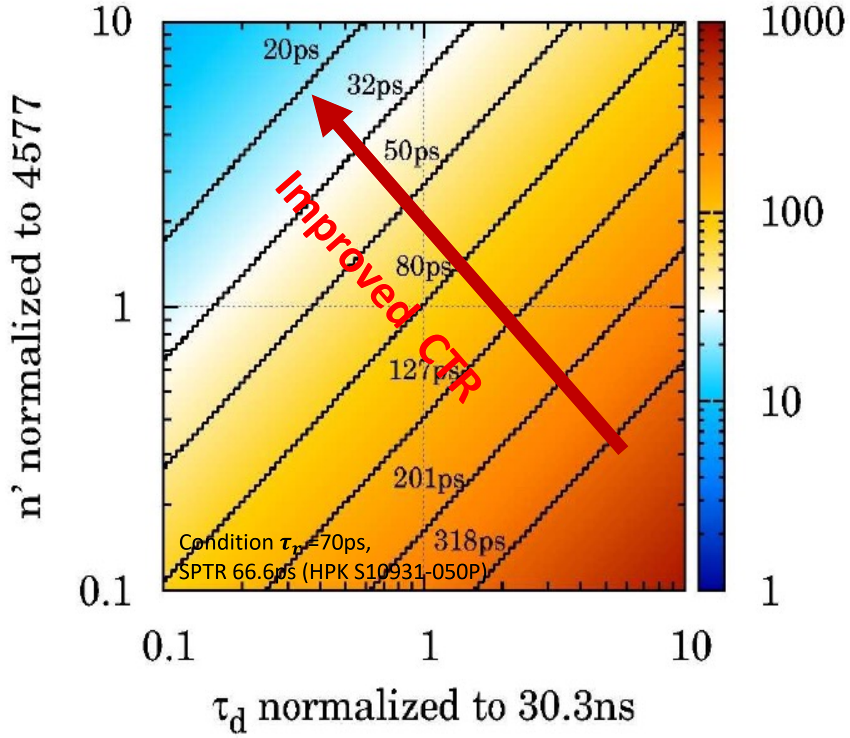
Main parameters influencing the time resolution of scintillating materials

Rise time(τ_r)



decay time (τ_d)

$$I(t) = Io(1 - e^{-t/\tau_r})e^{-t/\tau_d}$$

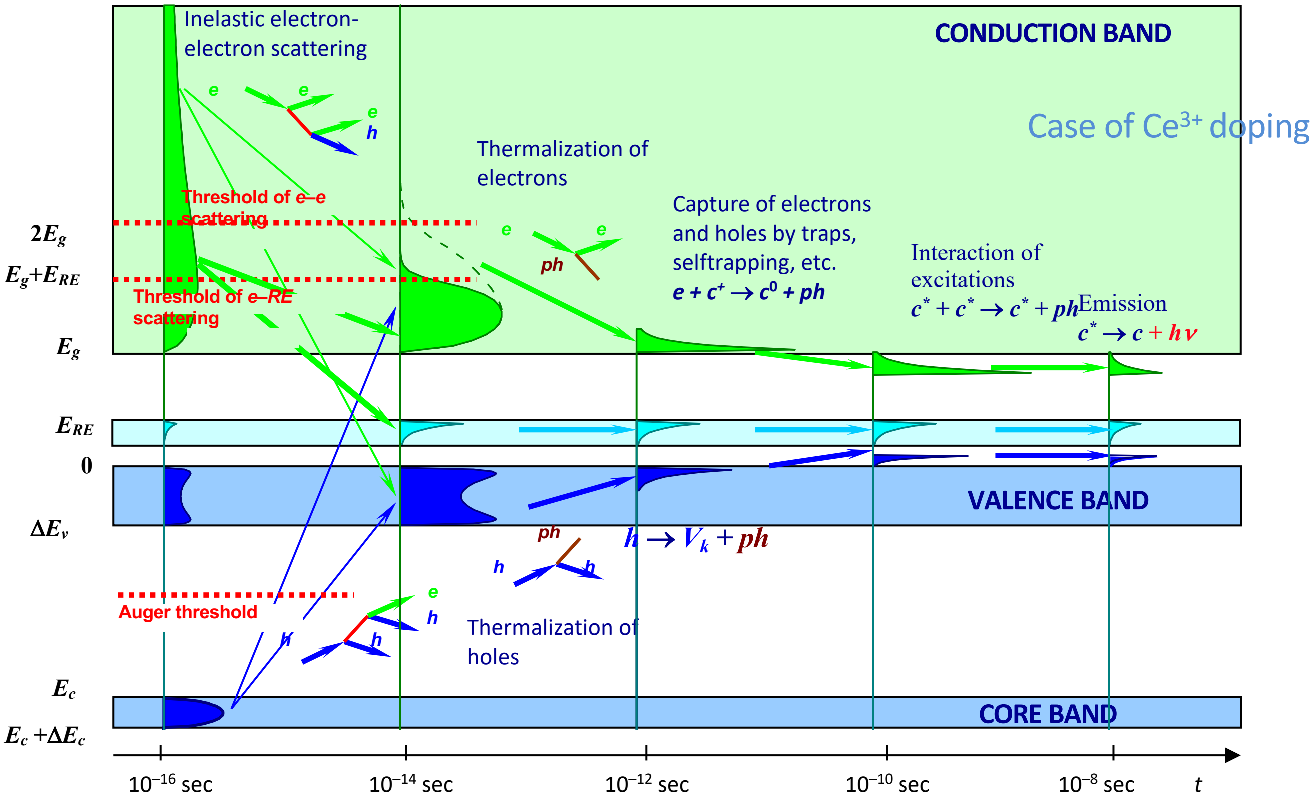


Coincidence time resolution $CTR \; \alpha \sqrt{\frac{\tau_d \tau_r}{N_{pe}}}$

S. Gundacker, PHD, CERN-THESIS-2014-034

Scintillation: a complex process chain

From eh pair creation to light emission



A. Vasiliev, Proceedings of The SCINT99 conference, Moscow, Faculty of Physics, Moscow State University, 2000, p. 43-52

Various possibilities for fast emission process

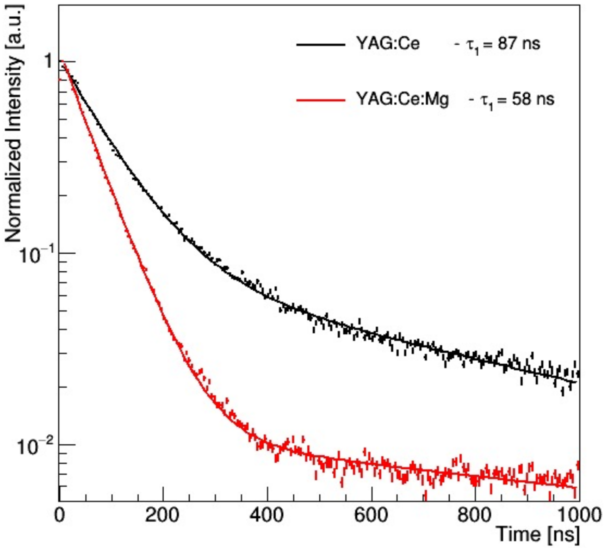
- Excitonic emission (STE, excitations of anion complexes)
- Emission of activators (Ce, Pr, …) Codoping:
- **Cherenkov radiation**
- **Crossluminescence**
- **Hot intraband luminescence (HIL)**
- **Quantum confinement driven luminescence:**

Slow

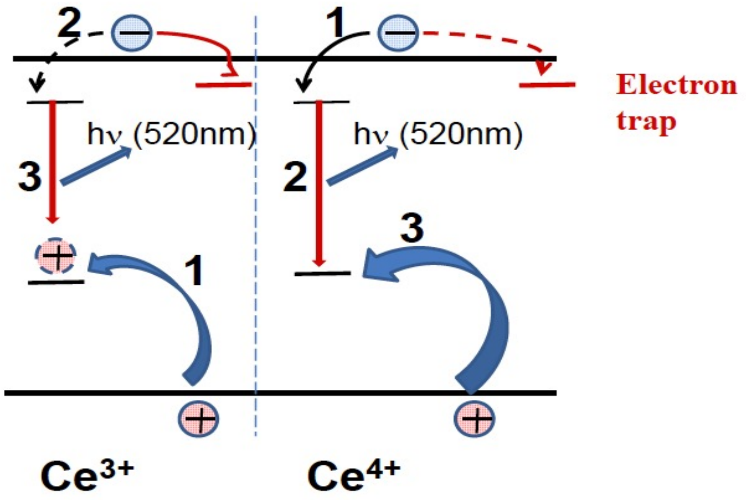
Ultra fast

Engineering of “standard” crystals
Example of codoping in Garnet

Decay time



Role of Ce^{4+}



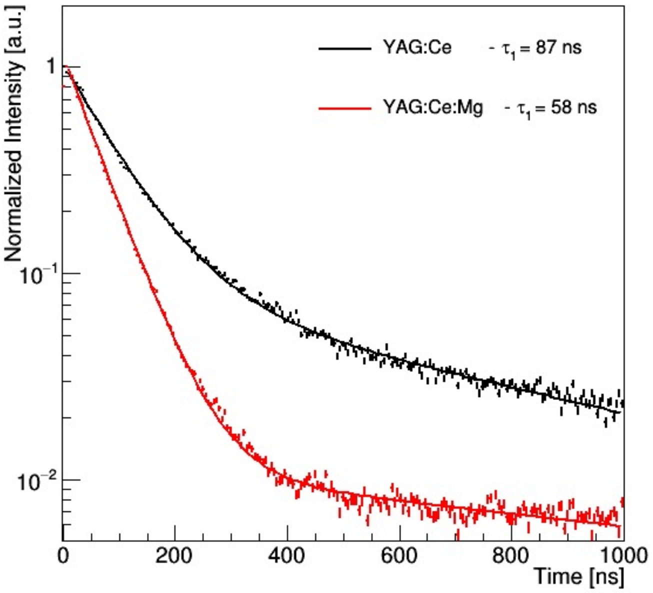
With codoping increase of Ce^{4+} => faster emission process

M. Lucchini et al, NIM A Volume 816 (2016), pp 176–183

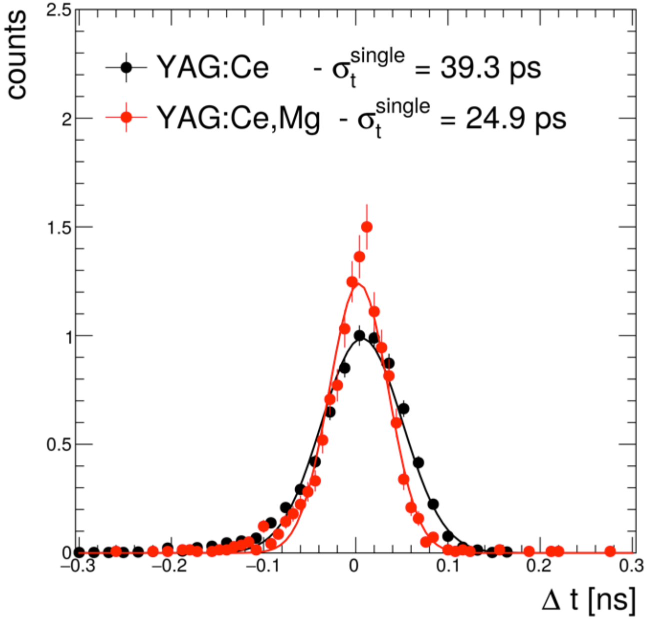
M. Nikl, A. Yoshikawa, Adv. Optical Mater. 2015, 3, 463–481
M. Nikl et al. Cryst. Growth Des. 2014 , 14 , 4827 .

Engineering of “standard” crystals
Example of codoping in Garnet

Decay time



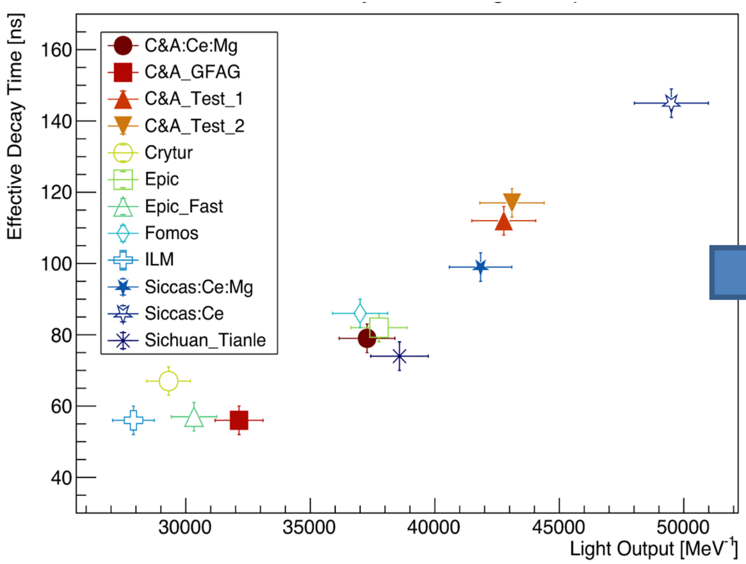
Time resolution with MIPs



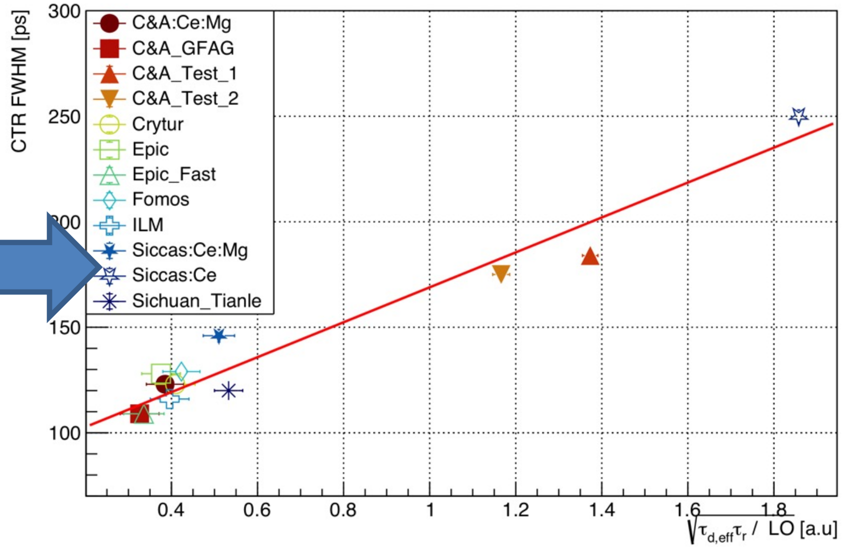
M. Lucchini et al, NIM A Volume 816 (2016), pp 176–183
M. Lucchini et al, NIM A Volume 852 (2017), pp 1.9,

Possibility to tune the timing properties

Effective decay time versus light ouptput for various GAGG samples



Time resolution @ 511 KeV versus photon density of various GAGG samples



Time resolution is inversely proportional to the photon time-density: $\sqrt{\frac{\tau_d \tau_r}{LO}}$

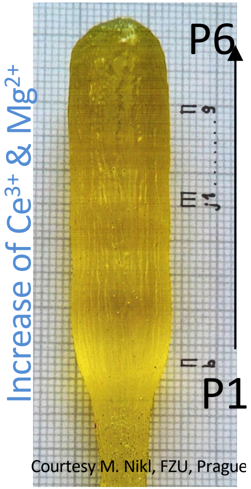
=> Light output loss is compensated by a decay time reduction

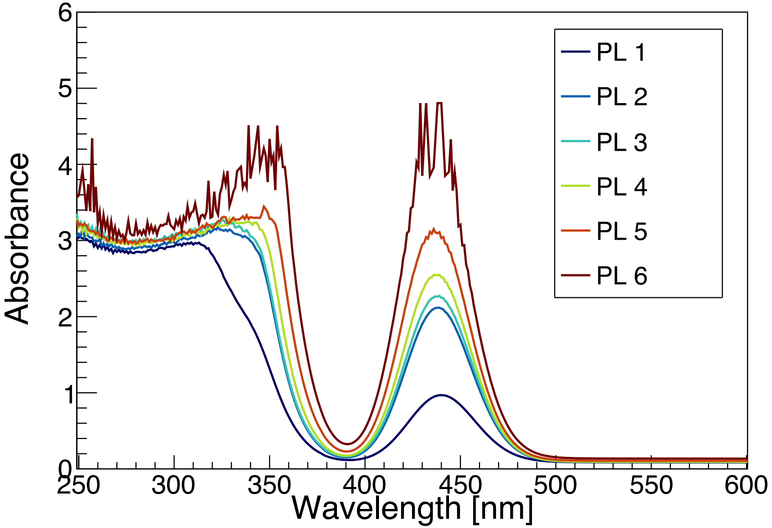
⇒Time resolution improves

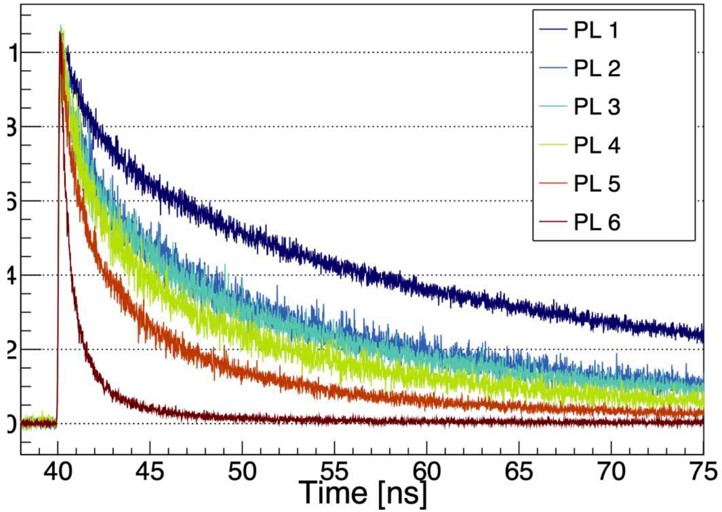
⇒Further effort to reduce decay time on going

L. Martinazzoli et al., NIM A, 1000 (2021), 165231

Development towards ultra fast GAGG:Ce by heavy Ce/Mg doping



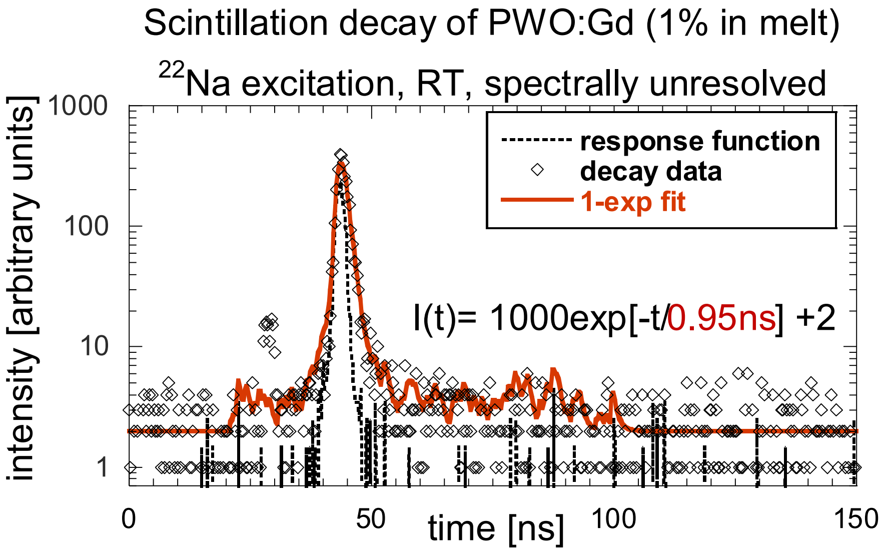


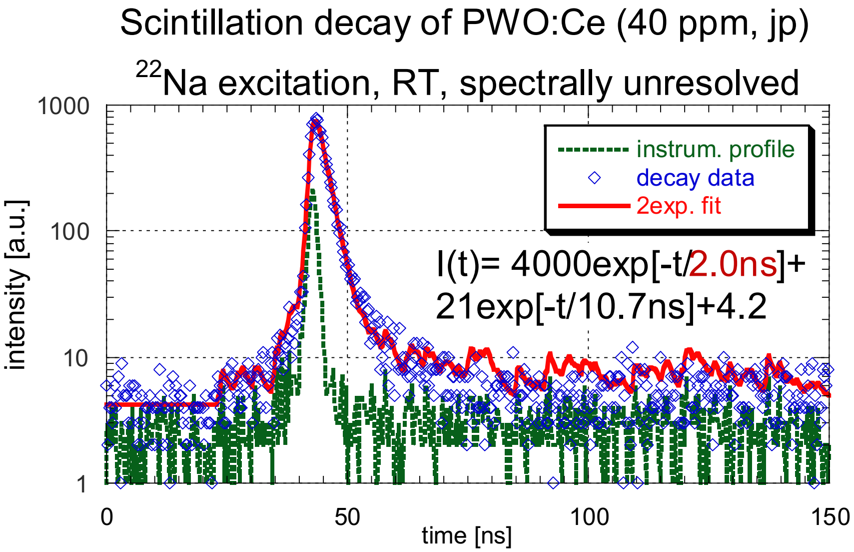


With increased concentration of Ce,Mg shortening of the decay down to **15ns**

L. Martinazolli et al, submitted to Light: Science & Applications

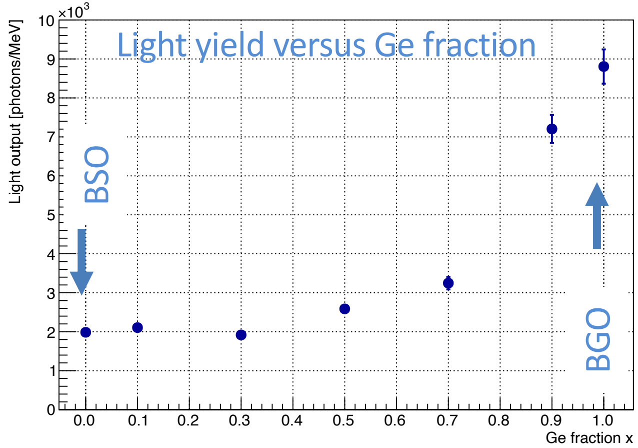
Towards very fast PWO

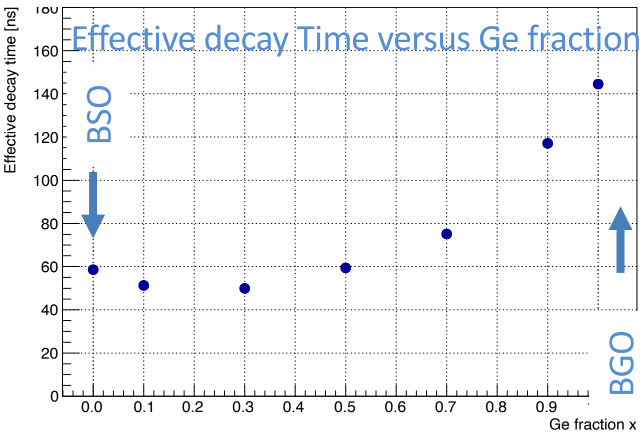




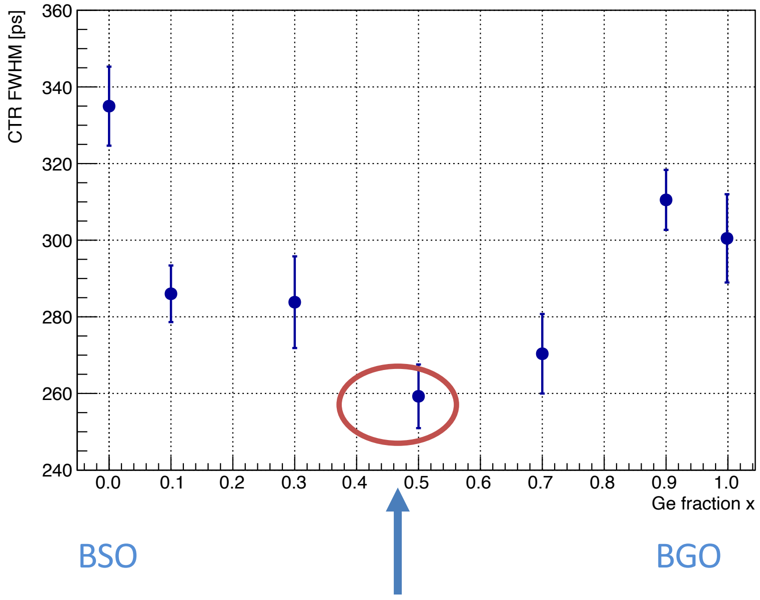
*M. Nikl et al, J.Cryst. Growth **229**, 312-315 (2001)*
*M. Nikl, et al, Radiation Measurements **33**, 705-708 (2001)*
*M. Kobayashi, et al: Nucl. Instr. Meth. in Phys. Res. A **459**, 482-493 (2001)*

Mixed Crystals BGO-BSO ($Bi_4(Ge_xSi_{1-x})_3O_{12}$)





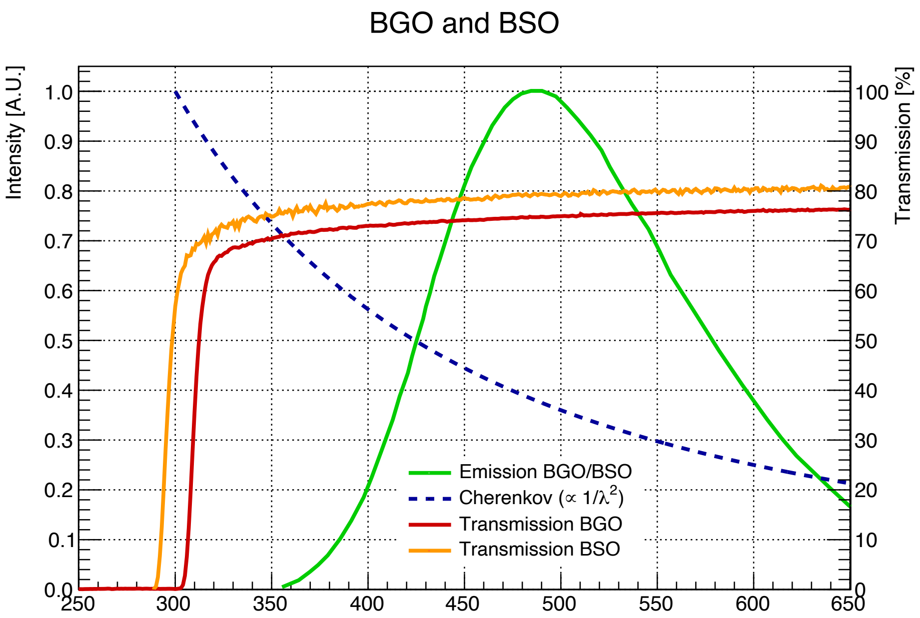
Coincidence time resolution @511Kev versus Ge fraction



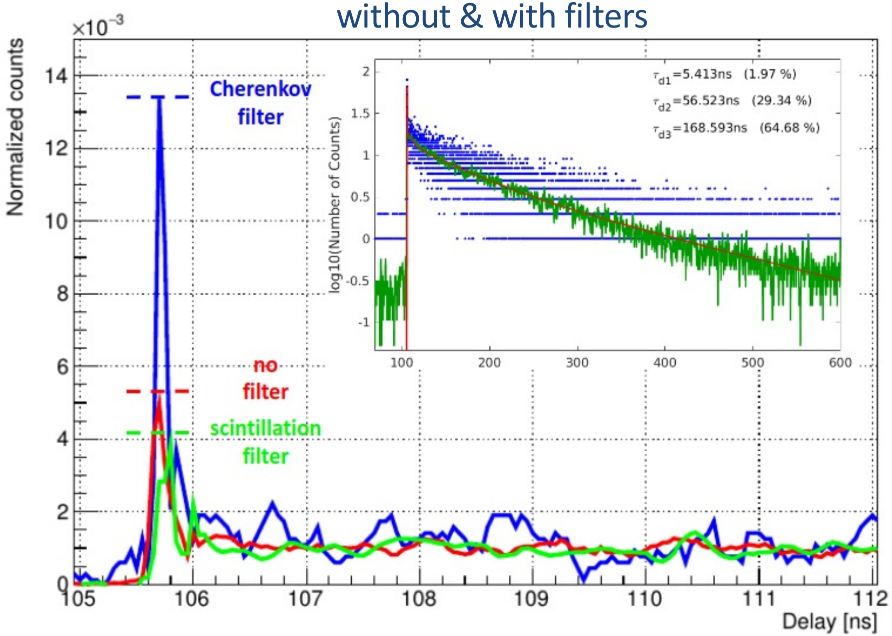
Optimal Ge fraction for time resolution

R. Cala et al, paper under review in NIMA

Exploitation of Cerenkov/scintillation in intrinsic scintillating crystals



Decay time spectra of BSO under 511 keV excitation without & with filters



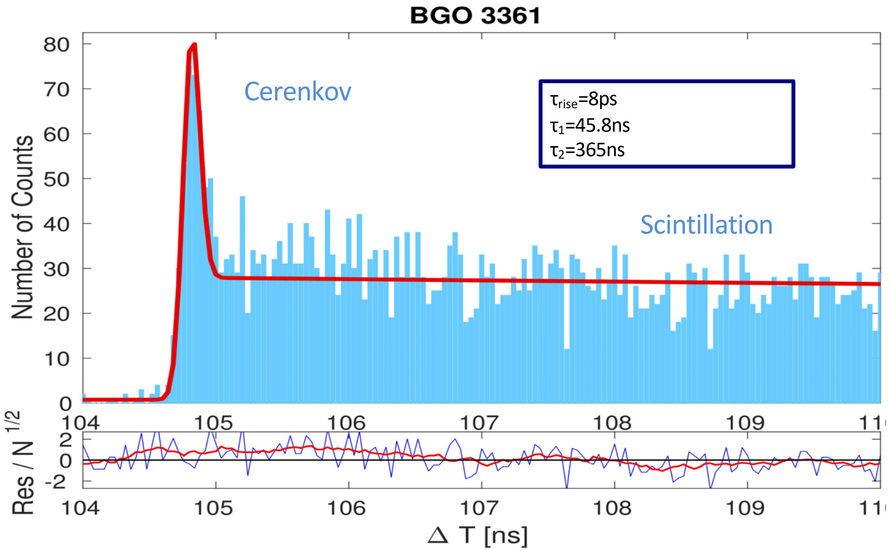
⇒ Possibility to separate Cerenkov from scintillation with filters &/or pulse discrimination

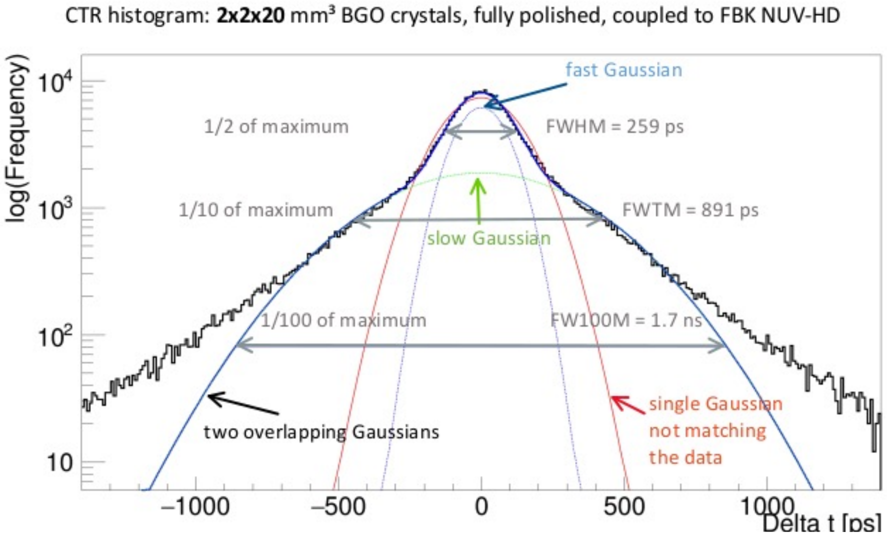
BSO (or mixed BGSO) is faster than BGO and has higher LY than PWO

⇒ Promising candidate for dual readout homogenous calorimeter

R. Cala et al, paper under review NIMA

Exploitation of Cerenkov to improve time resolution of BGO



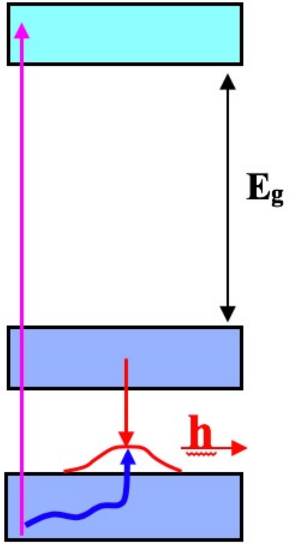


CTR:

S. Gundacker et al. (2019) Phys. Med. Biol. 64 055012
N. Kratochwil et al (2020), Phys. Med. Biol. 65 115004
N Kratochwil et al (2020) IEEE TRPMS 2020.3030483

Crossluminescence material

Radiative transition between the core- and valence bands.



Many possible materials

Compilation of CL data at 293 K

C.W.E. Van Eijk J of lum., Vol 6061, 1994936-941

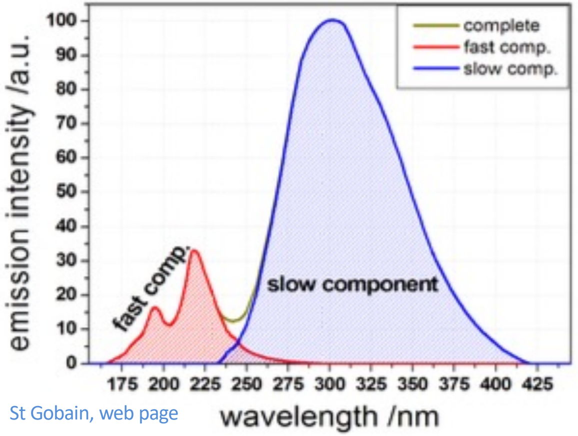
BaF$_2$ →

	$E(C-V)$ (eV)	$E(G)$ (eV)	Theoretical	Observed (eV)	λ (nm)	Light yield (photons/MeV)	τ (ns)	Density (g/cm^3)	References
KF	7.5–10.5	10.7	+	7.5–8.5	156	--		2.5	[13, 18]
KCl	10–13	8.4	–						
KBr	10–13	7.4	–						
KI	9.5–14	6.0	–	STE					
RbF	0–7.5	10.3	+	3–6	203, 234	1700	1.3	3.6	[11–14, 18]
RbCl	4–9	8.2	+	5.5–7.5	190	1		2.8	[12]
RbBr	6.7–9.5	7.4	/						
RbI	5–10	6.1	/	STE					
CsF	0–4.5	9.9	+	2.5–4	390	2000	2.9	4.1	[6, 11, 14]
CsCl	1–5	8.3	+	4–5.5	240, 270	900	0.9	4.0	[6, 14, 15, 17, 18]
CsBr	4–6	7.3	+	4.5–6.5	250	20	0.07	4.4	[6, 14, 15, 18]
CsI	0–7	6.2	/	–/STE					
CaF_2	12.5–17.3	12.6	–	–/STE					[1]
SrF_2	8.4–12.8	11.1	/	–/STE					[1]
BaF_2	4.4–7.8	10.5	+	5–7	195, 220	1400	0.8	4.9	[1, 3, 4, 9]
$K_xRb_{1-x}F$				5–6/8					[13, 18]
$KMgF_3$				6–9	140–190	1400	1.3	3.2	[7–10]
$KCaF_3$				6–9	140–190	1400	< 2	3.0	[10]
KYF_4					170	1000	1.9	3.6	[9, 16]
K_2YF_5				5.5–8.5	170	300	1.3	3.1	[8, 9]
$KLuF_4$				5.5–8.5	170–200	~ 200	1.3	5.2	[8, 9, 16]
KLu_2F_7				5.5–8.5	165	~ 200	< 2	7.5	[8]
K_2SiF_6				5–9	140–250				[21]
$CsCaCl_3$					250, 305	1400	~ 1	2.9	[10, 17, 19]
$CsSrCl_3$					260, 300		~ 1		[19, 21]
$LiBaF_3$					190, 230	1400	0.8	5.2	[10]
$BaMgF_4$					190, 220	1000		4.5	[21]
BaY_2F_8				4–7.5			0.9	5.0	[20]
K_2LiGaF_6				5–9	140–250				[21]
K_2NaAlF_6				5–9	140–250				[21]

Very fast emission < 2ns but emission < 300nm

Suppression of slow component in BaF_2

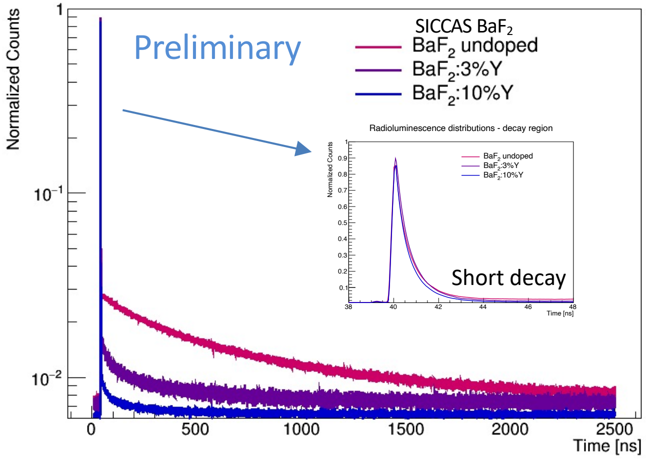
BaF_2 emisison spectra



St Gobain, web page

Sub ns emission but in UV
& additional slow component

Decay time spectra for various % Y doping



R. Cala et al, CERN EP_CMX

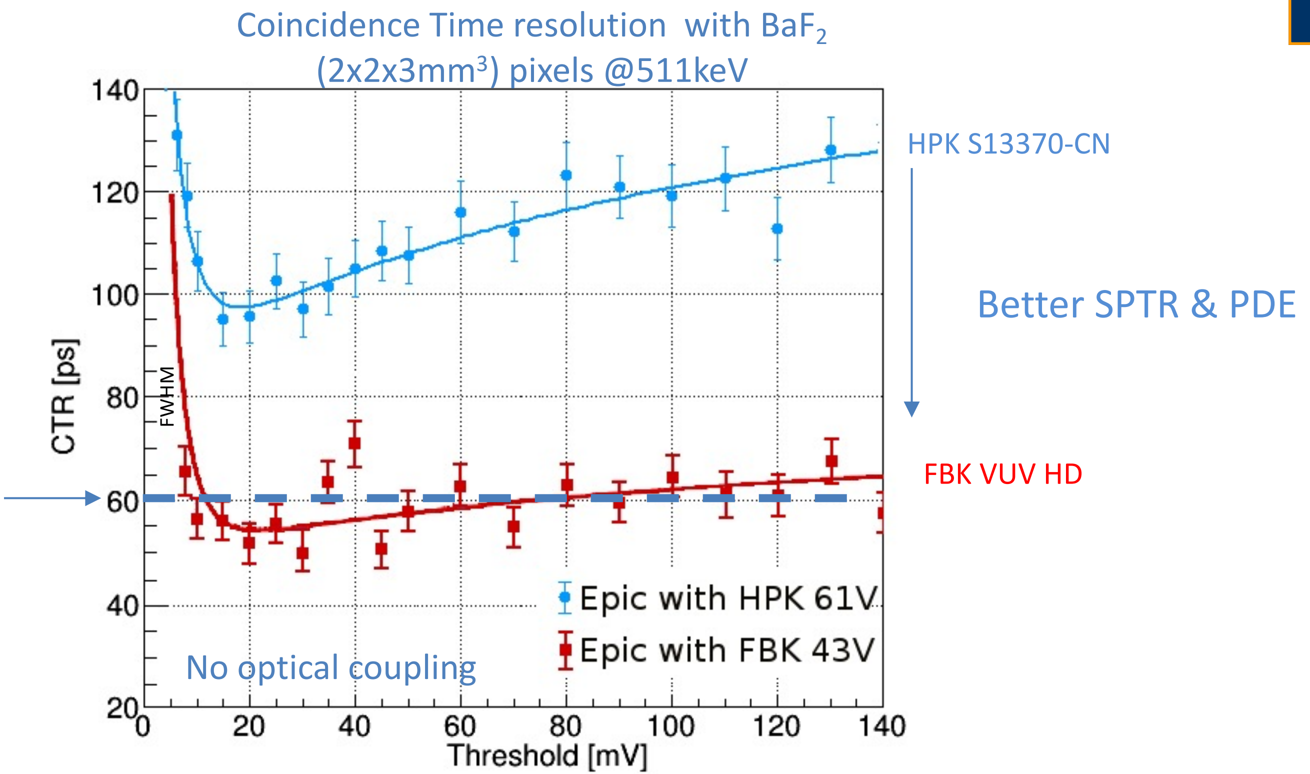
R&D to suppress the slow component by doping

⇒ No change in short decay

⇒ but slow component suppression

J. Chen, et al., IEEE Trans. Nucl. Sci., vol. 65, no. 8, pp. 2147-2151, 2018.
S. Gundacker et al., Phys. Med. Biol. 66 (2021) 114002

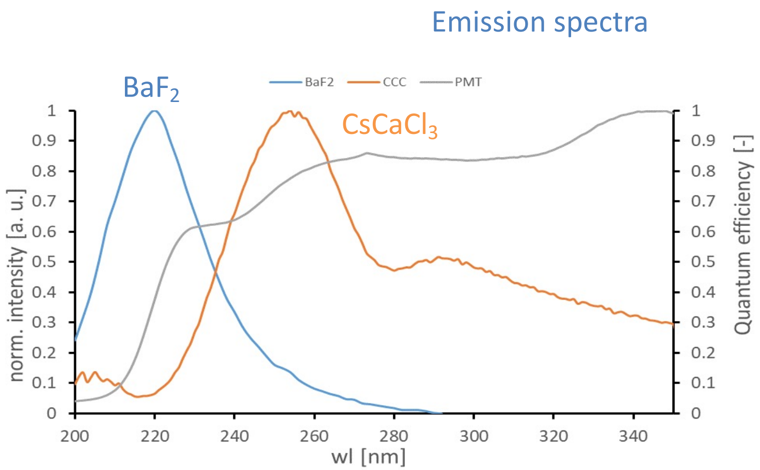
Improvement of UV photodetection



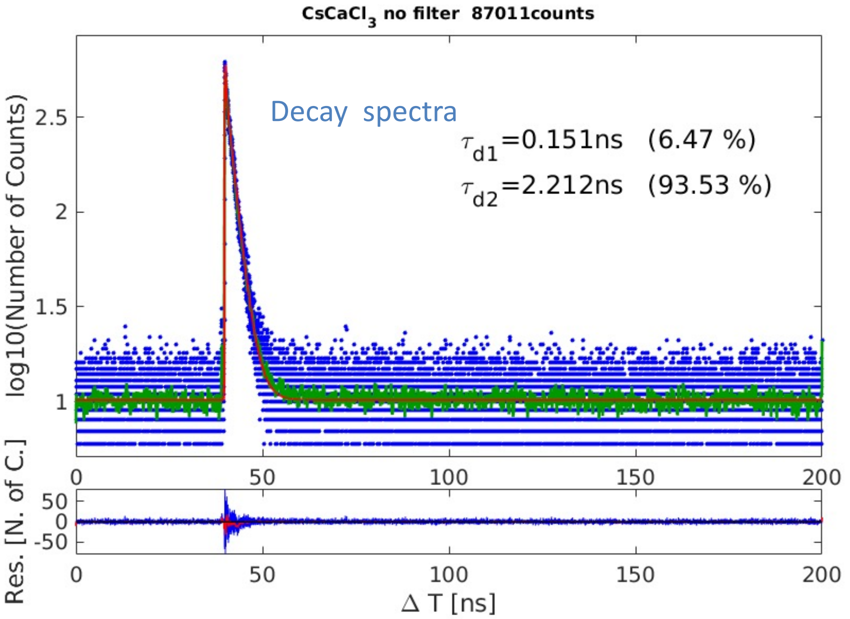
Further improvement of PDE in UV and optical coupling may improve time resolution of BaF_2 toward 30ps

R. Pots et al, Front. Phys. | doi: 10.3389/fphy.2020.592875
S. Gundacker et al., Phys. Med. Biol. 66 (2021) 114002

Development of cross luminescence material more in UV visible region



Courtesy V. Vanecek, M. Nikl, FZU Prague
Data for BaF_2 from M. Laval et al. , NIM Phys. Res., 206 (1983) 169–176



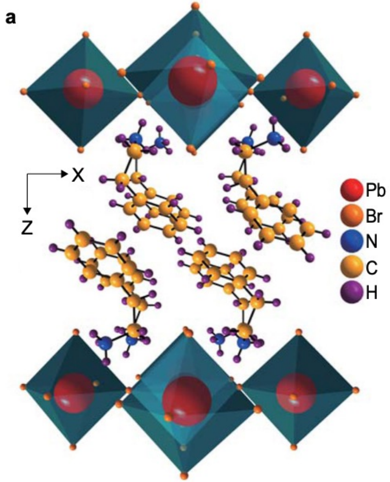
$CsCaCl_3$:2 emissions @ 260nm & 290nm
2 fast decay times: 0.15ns, 2.2ns

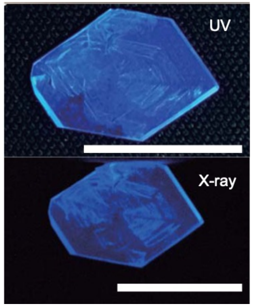
FZU Fyzikální ústav Akademie věd České republiky

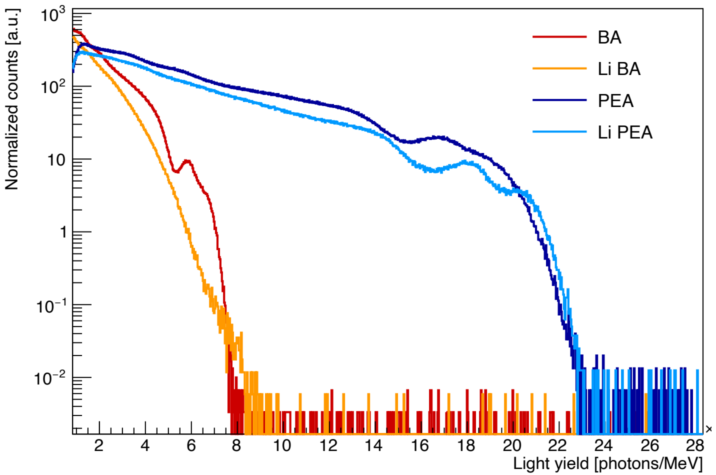
V. Vanecek et al., Optical Materials X 12 (2021) 100103

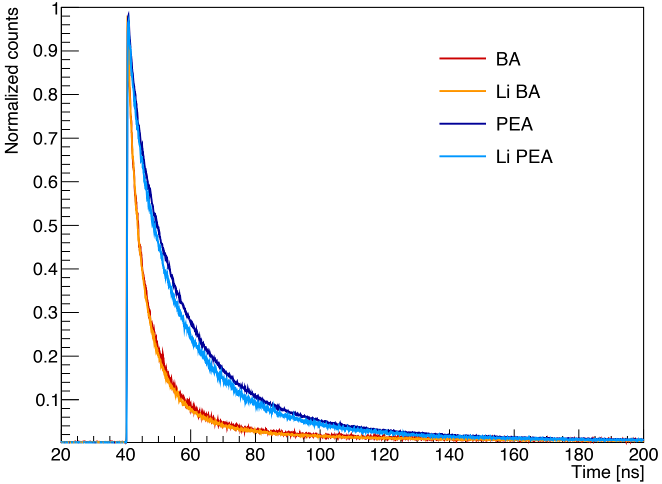
Two dimensional Hybrid perovskites

An organic-inorganic hybrid structure.









Relative high light output 20000ph/MeV For PEA type

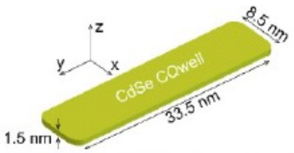
Fast decay time < 20ns

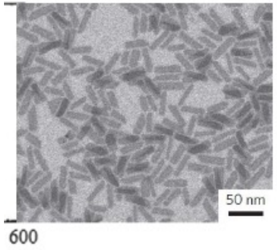
A. Xie et al, Chem. Mater. 2020, 32, 8530–8539

R. Cala et al, to be submitted to ACS Photonics

CdSe quantum well/quantum dot

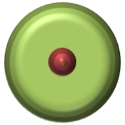
CdSe nanoplatelet

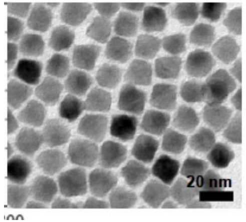




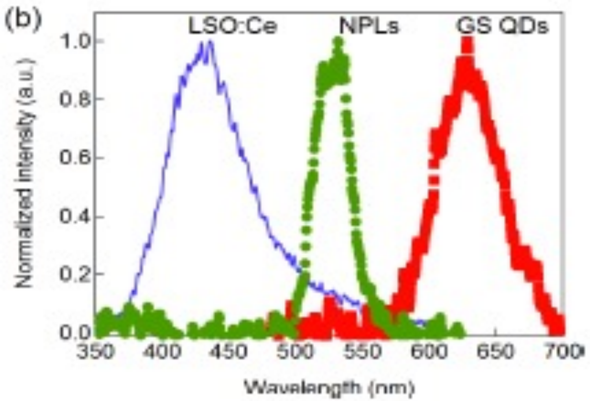
J.Q. Grim et al., Nature Nanotechnol. 9 (2014) 891.

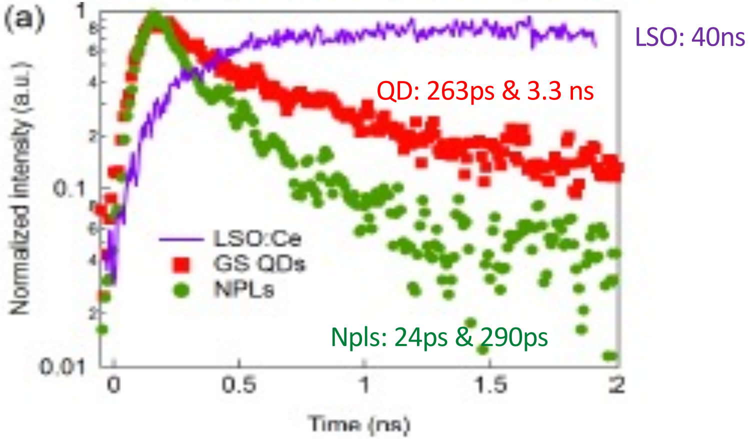
CdSe/CdS Giant shell Quantum dot





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

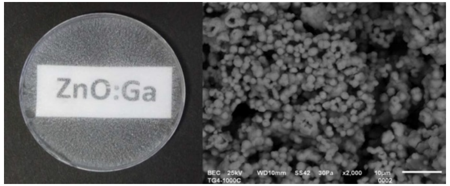




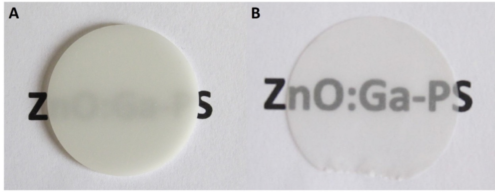
R. Martinez Turtos et al., 2016 JINST 11 (10) P10015

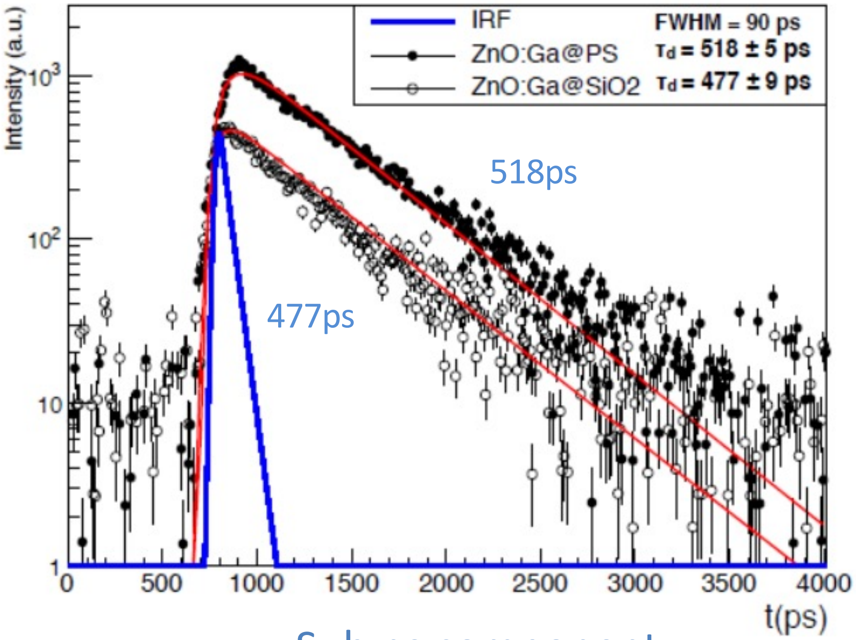
ZnO:Ga nanomaterial

In SiO_2



In Polystyrene





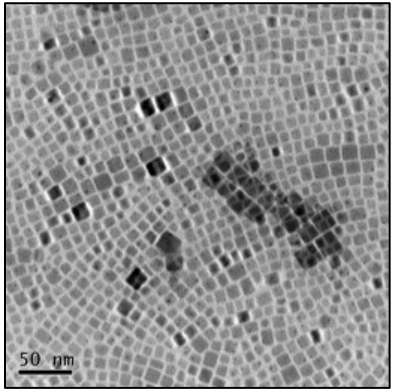
Sub ns component

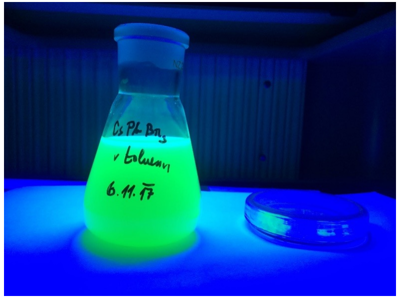
Procházková et al., Radiat Meas 90, 2016, 59-63
*Buresova et al, Opt. Express **24**, 15289 (2016)*

R. Turtos et al, Phys. Status Solidi RRL 10, No. 11, 843–847 (2016) /

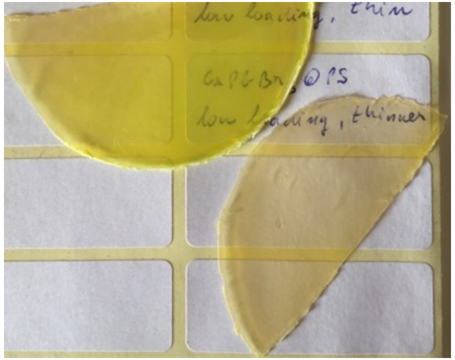
Perovskite nanomaterials

$CsPbBr_3$ nanocrystals



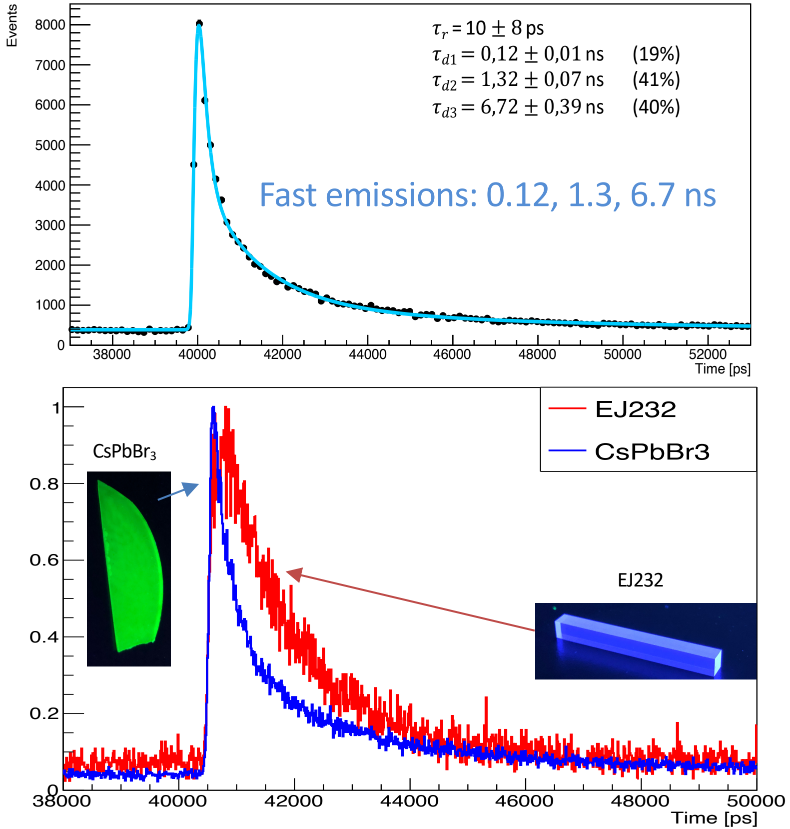


$CsPbBr_3$ nanocrystals imbedded in polystyrene



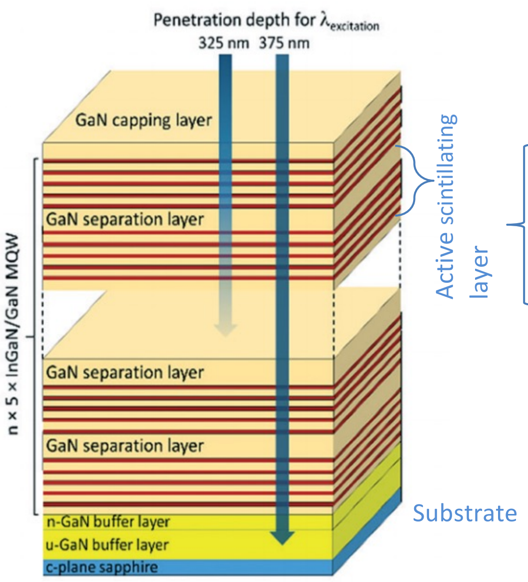
Courtesy V. Čuba, K. Děcká, A. Suchá CTU, Prague

X ray decay time



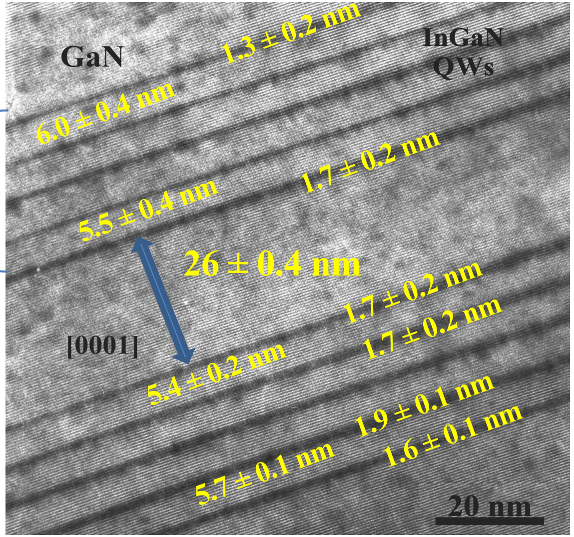
F. Pagano et al. , CERN, CTU

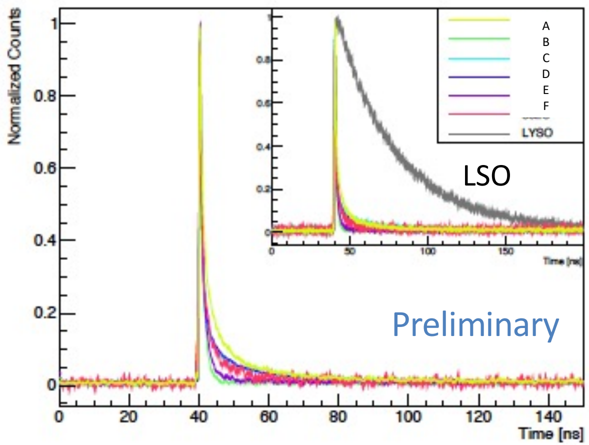
InGaN/GaN heterostructure: Multiple Quantum Wells (MQW)



T. Hubacek, CrystEngComm, 2019, 21, 356

Picture from A. Hospodkova

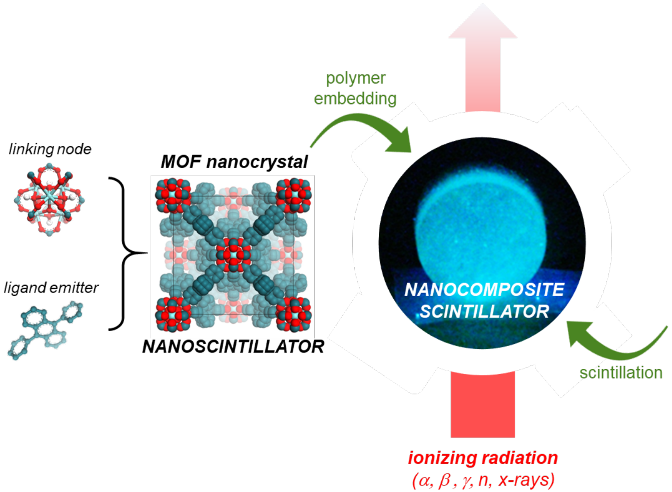




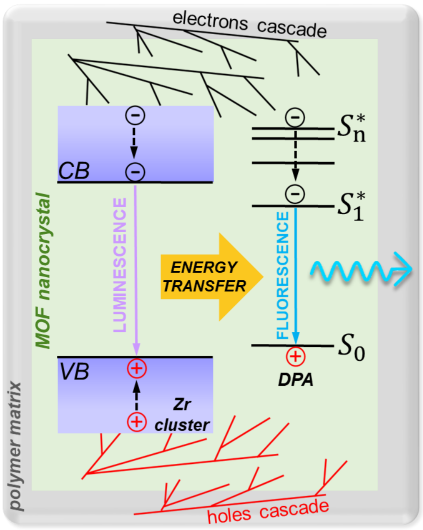
F.Pagano, et al, CERN &FZU

Sub-ns fast emisison

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

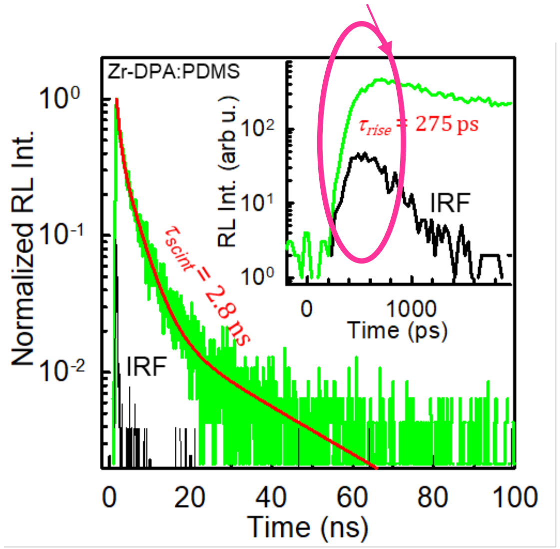


Sensitized Scintillation



Rise time limited by the instrumental response function

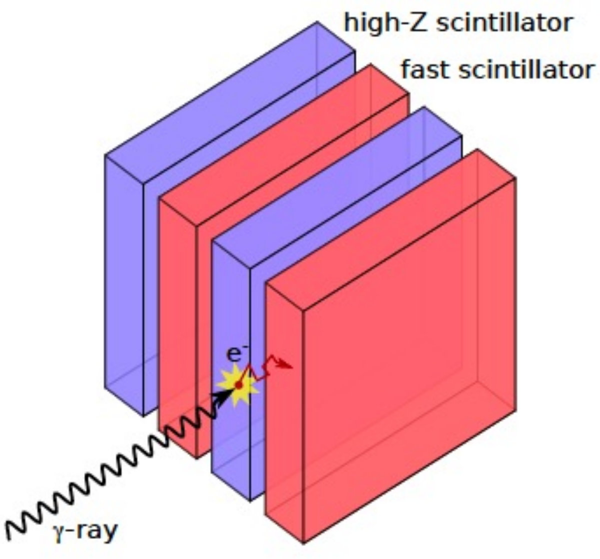
REAL $\tau_{rise} << 300$ ps

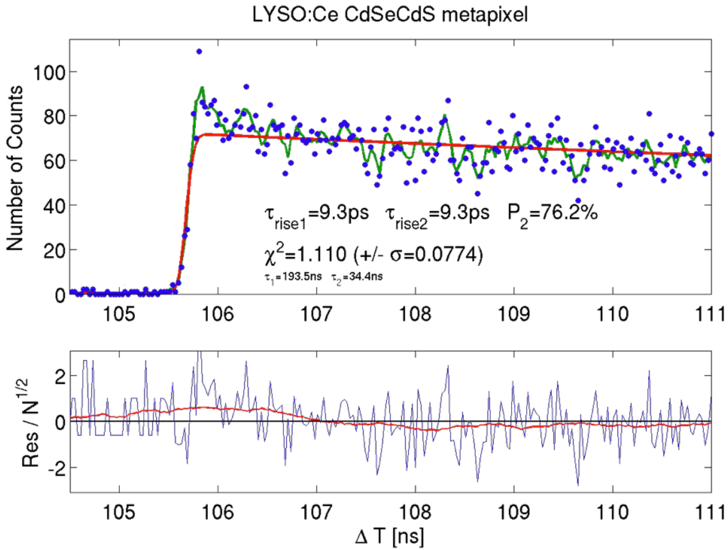


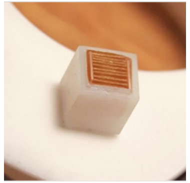
J. Perego, et al. Nat. Photonics (2021). https://doi.org/10.1038/s41566-021-00769-z

Heterostructure concept

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

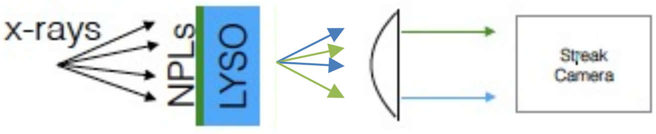


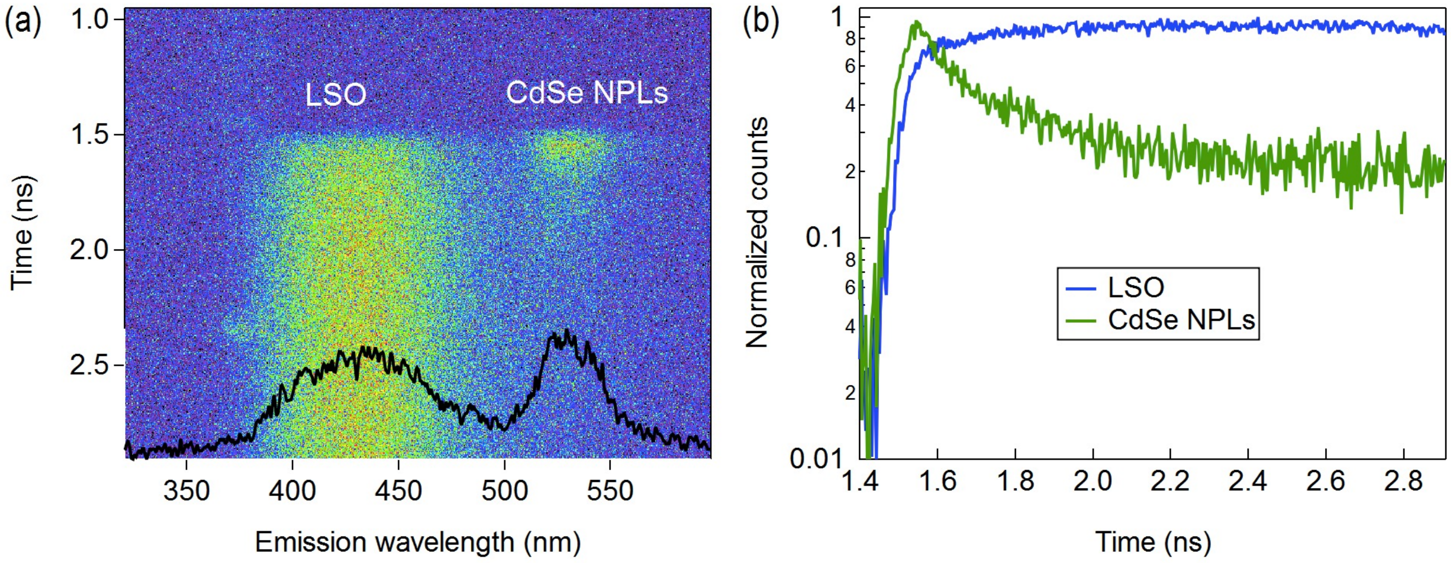




ERC grant 338953 (TICAL , PI P. Lecoq)
R. M. Turtos et al, Phys. Med. Biol. 64 (2019) 85018
F. Pagano et al, Submitted to PMB

CdSe nanoplatelets deposited on LYSO

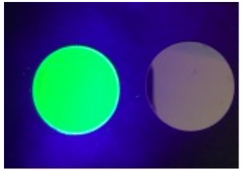


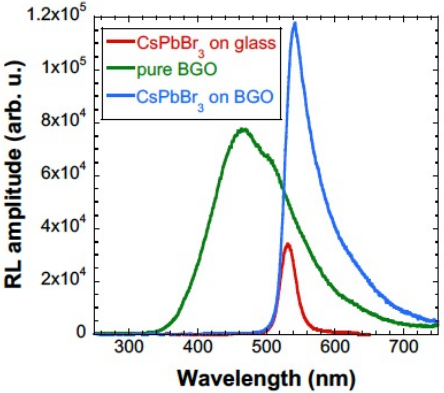


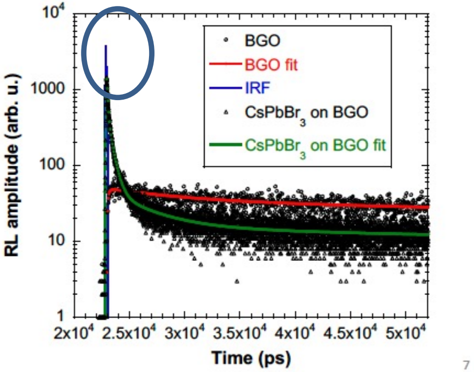
R. Martinez Turtos et al., JINST_068P_06

$CsPbBr_3$ nanocrystals deposited on BGO and GAGG

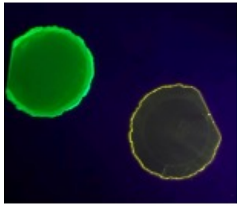
BGO

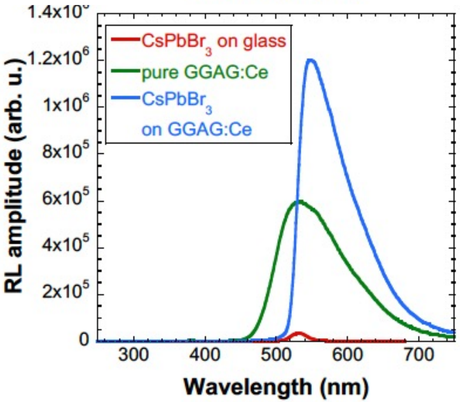


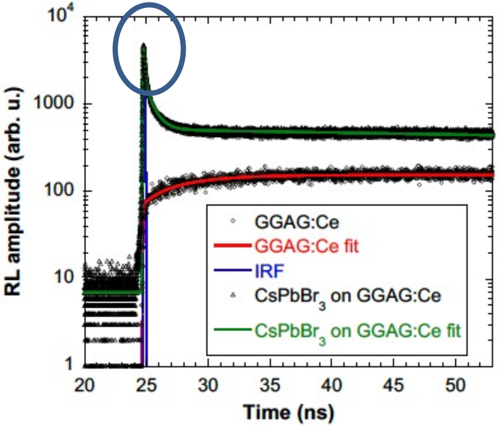




GAGG



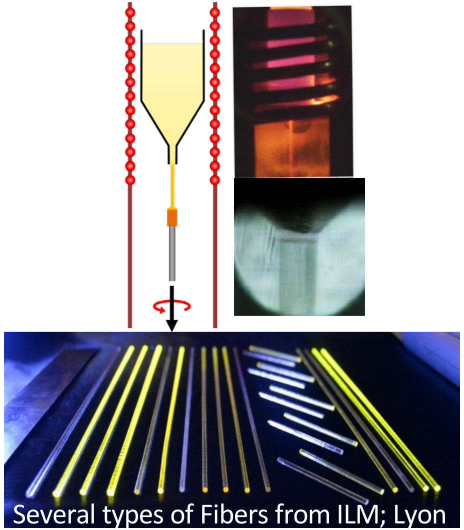




K. Děcká et al., Nanomaterials 2022, 12, 14. https://doi.org/10.3390/nano12010014

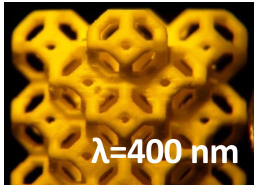
Future detector concept: Benefit from new developments

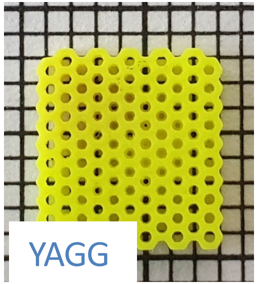
Micropulling down technique



3D printing

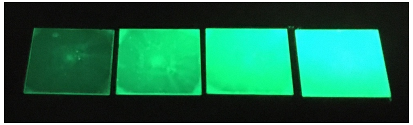
YAG





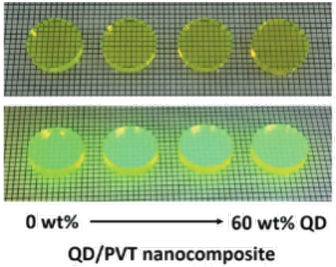
Courtesy of G. Dossovitky, Kurchatov Institute

$CsPbBr_3$ Nano crystals thin films deposited on glass substrate



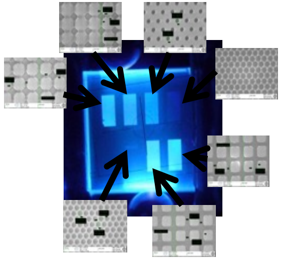
Courtesy V. Čuba, K. Děcká, CTU, Prague

CdxZn1-xS/ZnS (CZS) QD composite

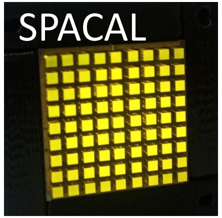


C. Liu *et al. ACS Nano, 2017*

Photonic Crystals



A. Knapitsch et al. IEEE TNS, VOL. 63, NO. 2, April 2016
M. Salomini et al., Crystals 2018, 8(2), 78;



Conclusion

Many progress in understanding key parameters for fast timing detectors have been made

Many approaches in the scintillation field have been explored and need R&D to be pursued:

- Develop bright and fast scintillator:
  - Search for new material
  - Band gap engineering
- Exploit better: cross luminescence and Cerenkov emission
  - Request for better UV sensitive photodetector and optical glue
- Research of intraband luminescence material
- Research of for fast nanomaterials with bright sub-ns light emission based on quantum confinement

Together with new developments in:

- Production methods
- Photonic crystals, plasmonic effect to manipulate and enhance the light output

=> Open perspectives for innovative concepts of future detectors with multi-functionalities

Acknowledgement

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