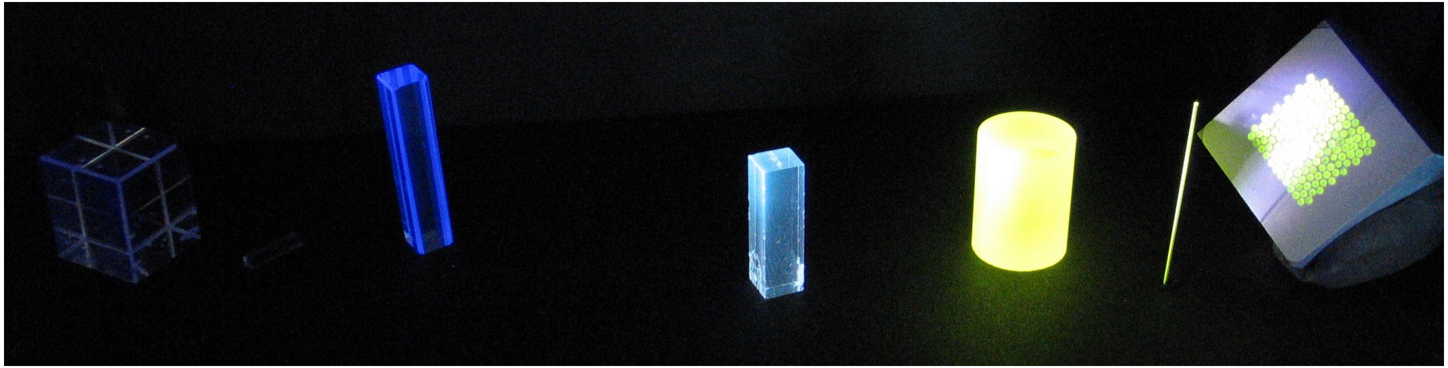
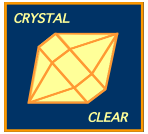
RECENT DEVELOPMENTS IN THE FIELD OF SCINTILLATORS FOR RADIATION DETECTORS

E. Auffray, *CERN, EP-CMX*

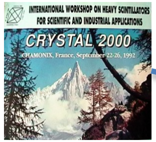


120 years of inorganic scintillators

1990: Request for scintillators for LHC
Birth of SCINT community



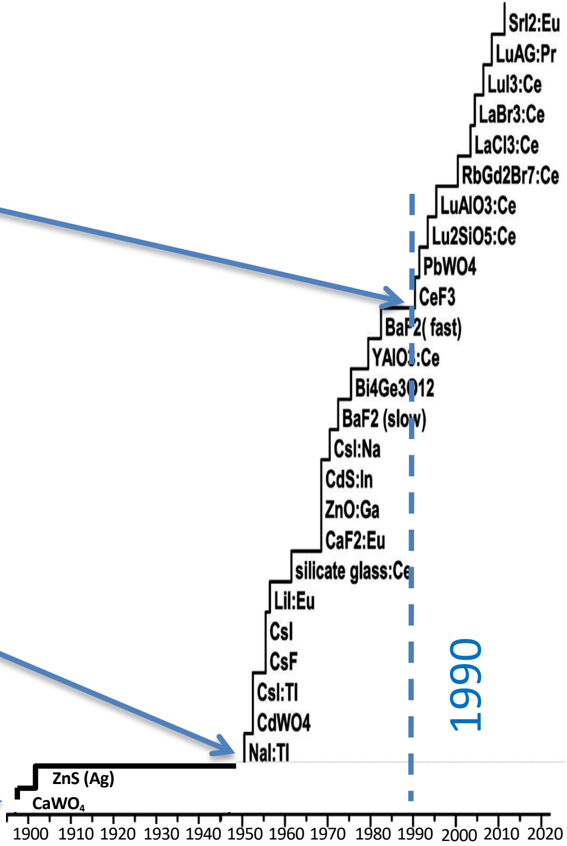
1991



1992
1[st] Scint conference

1930-1940:
Invention of the photomultiplier tube

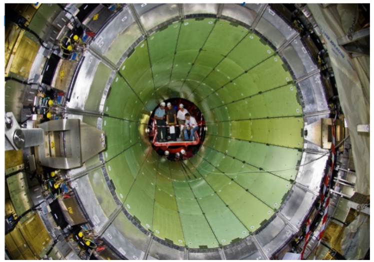
1896:
CaW04 used by Roentgen



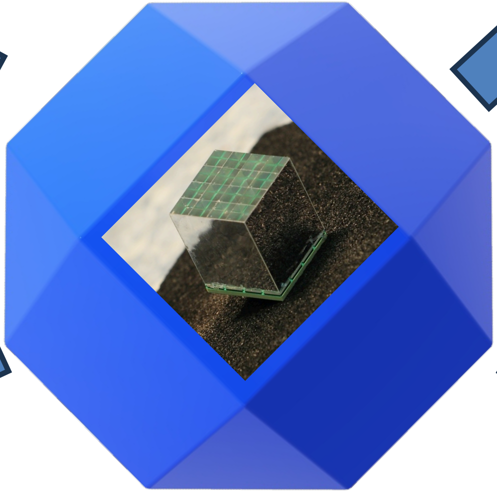
P. Dorenbos, Optical Materials: X 1 (2019) 100021,

A wide range of applications using scintillators

High Energy Physic

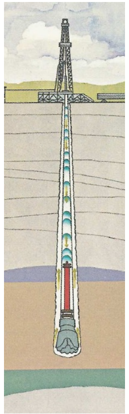


Medical applications



security

Oil well logging



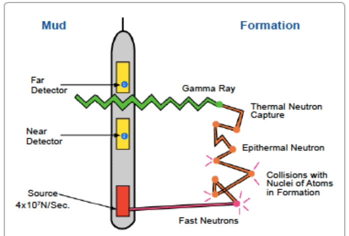


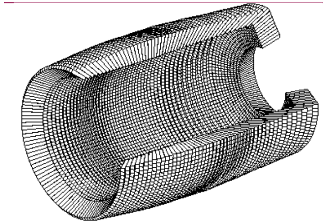
Figure 6: Generalized Neutron Logging Tool illustrates a typical neutron logging tool (Schlumberger, 2010).

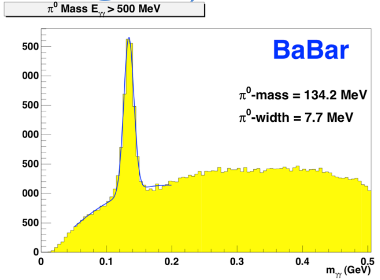
A. Bu, Oil Gas Res 2016, 2:2
DOI: 10.4172/2472-0518.1000113

Many other applications

Inorganic scintillators in HEP

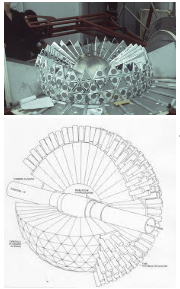
6580 CsI:Tl crystals:Babar @SLAC, 1999

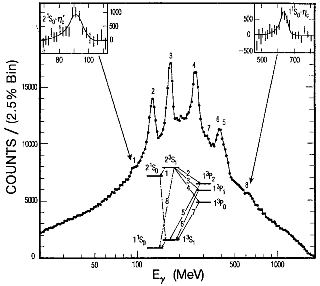


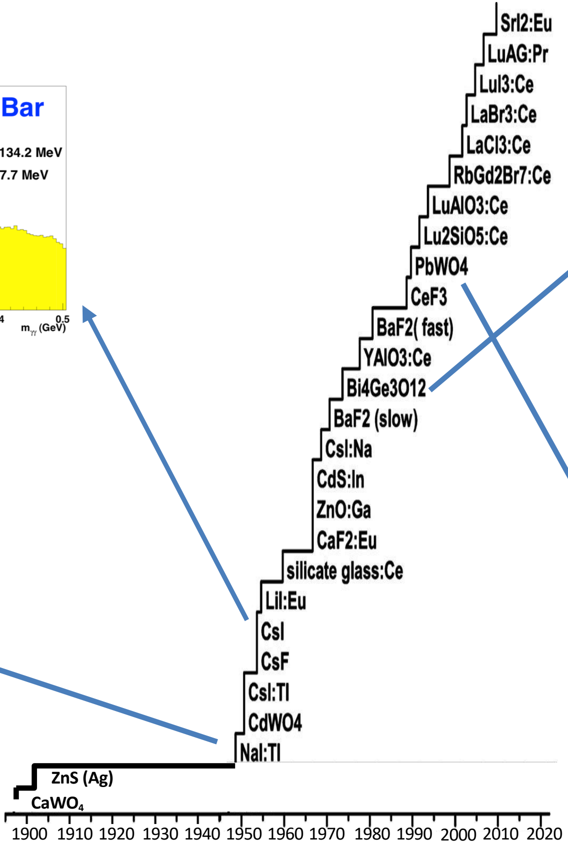


642 NaI (Tl): Crystal Ball @SLAC, 1979

Charmonium spectra

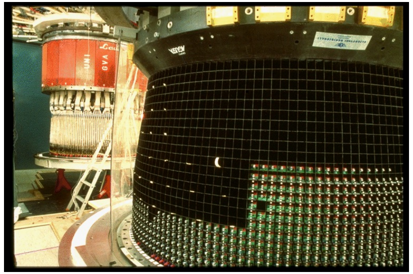




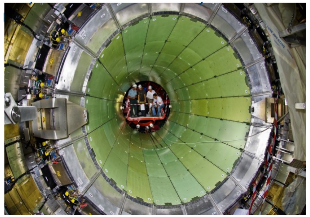


P.Dorenbos,, 10.1109/TNS.2009.2031140

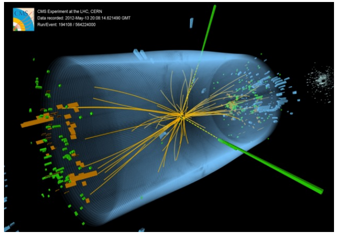
10752 BGO: L3 calorimeter @LEP 1989

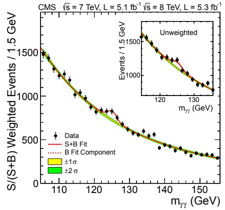


75848 PWO: CMS calorimeter @LHC 2008



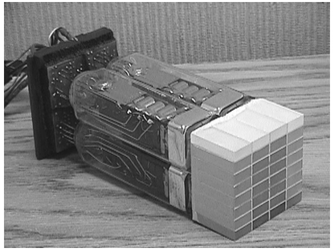
Higgs discovery in 2012

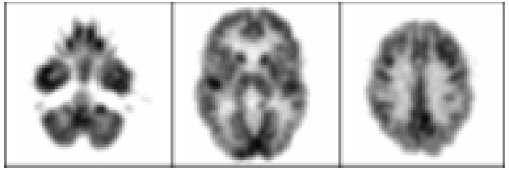




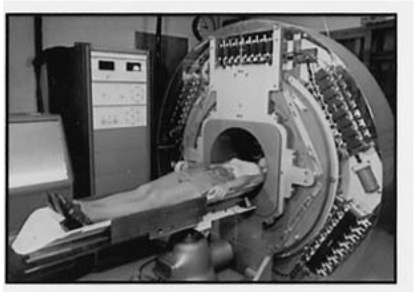
Inorganic scintillators in PET imaging

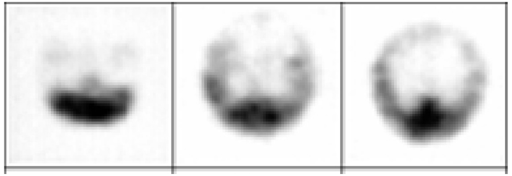
4096 BGO: ECAT 931 1985



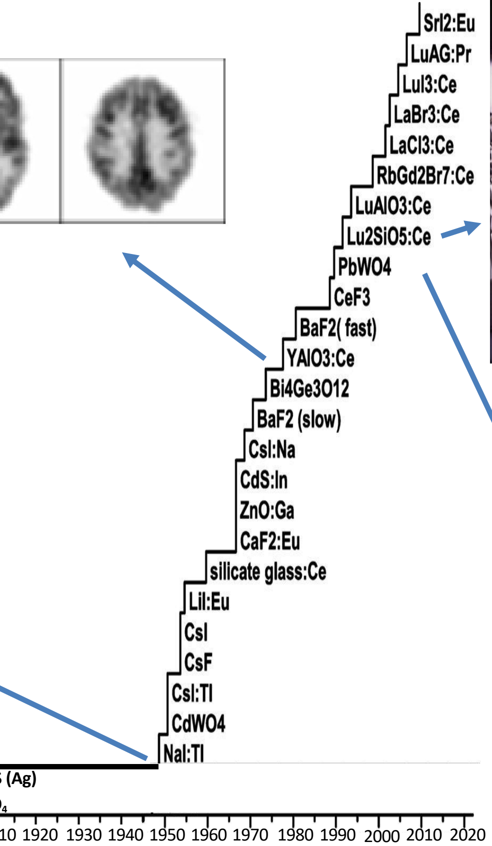


48 NaI (Tl): PETIII, 1974

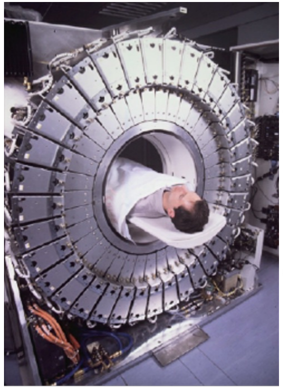




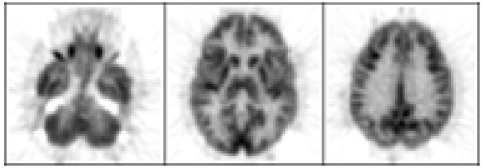
From Uwe Heinrichs forschungszentrum Julich



P.Dorenbos,, 10.1109/TNS.2009.2031140

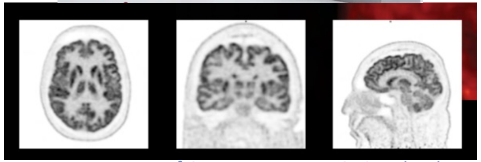


1995 LSO: ECAT EXACT HR+



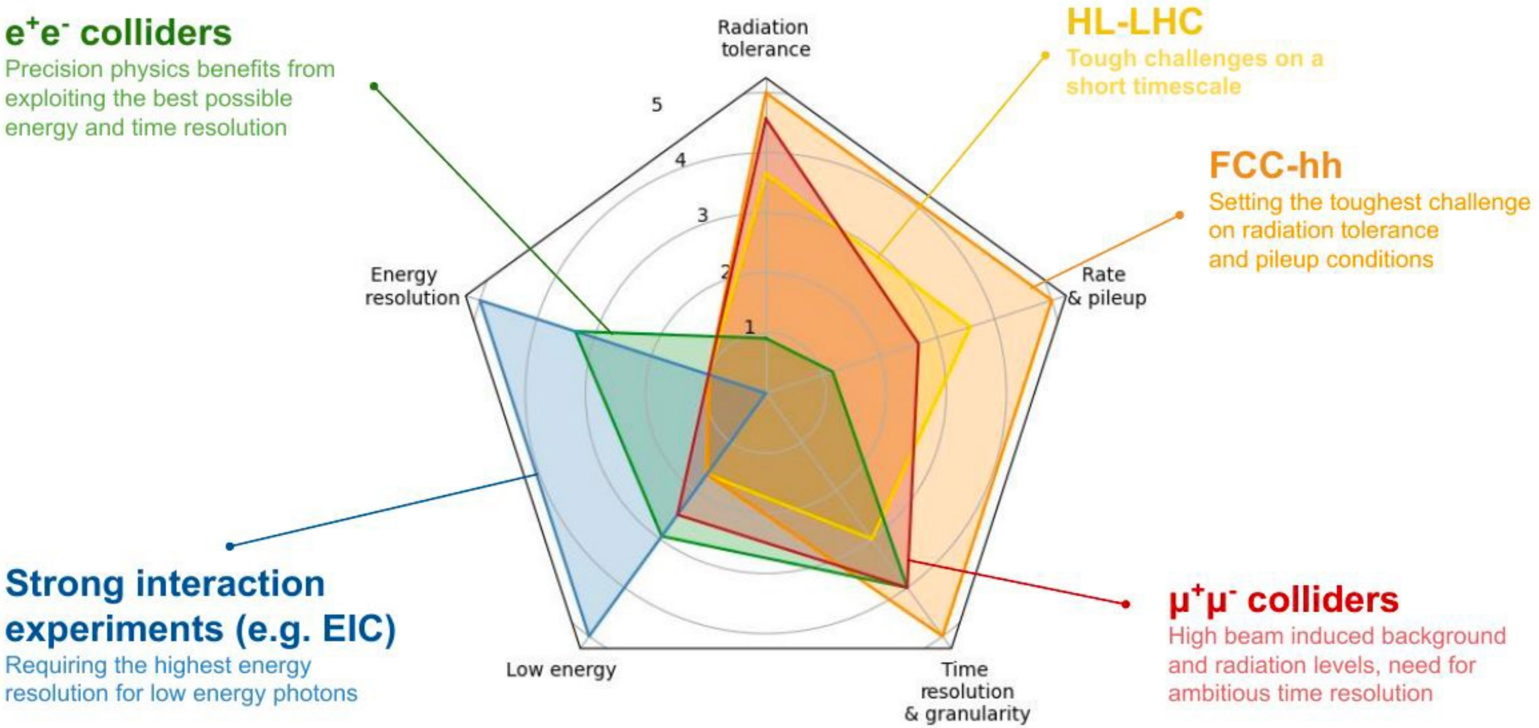
2018 LSO: TOF PET Biograph Siemens

From SIEMENS Healthcare web page



Data courtesy of CHUV, Lausanne, Switzerland

A large variety of future detectors with different requirements



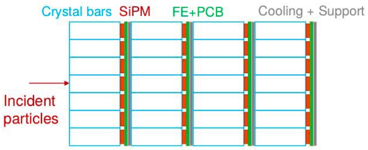
M.T. Lucchini, Scint2022 conference

A large variety of proposals with inorganic scintillators

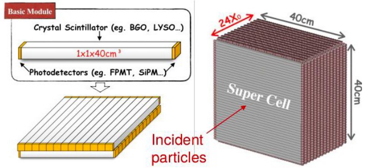
e^-e^+ colliders

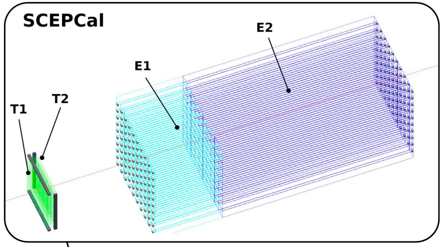
CEPC

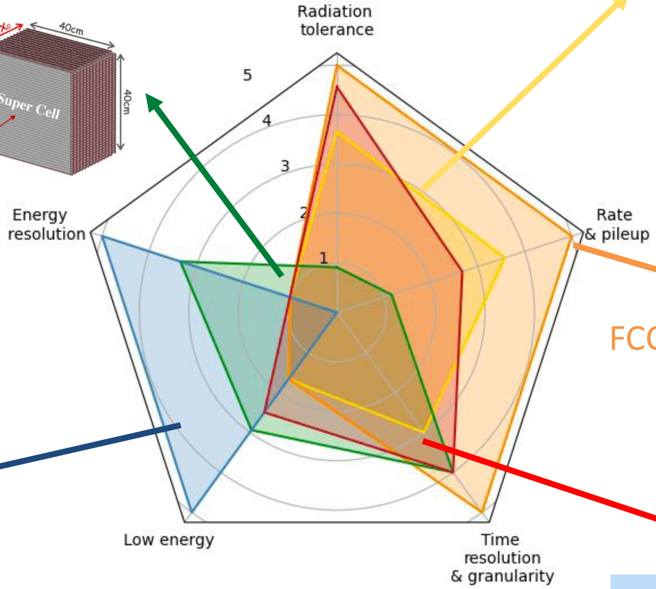
Design 1



Design 2

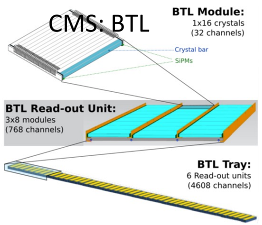




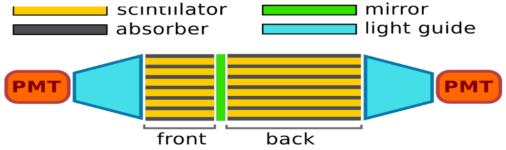


HL-LHC

CMS: BTL

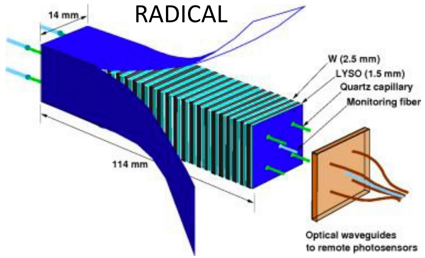


LHCb : SPACAL

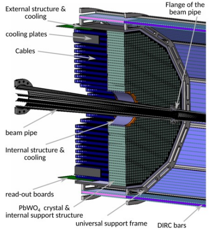


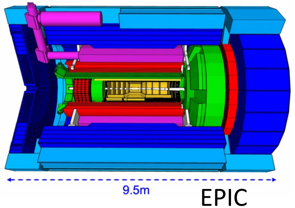
FCC-hh

RADICAL



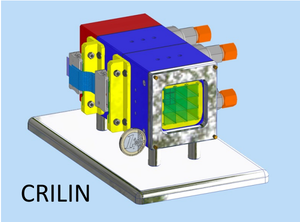
Strong interaction experiments (eg EIC)





EPIC

$\mu^-\mu^+$ colliders



CRILIN

Fast timing ever increasing request

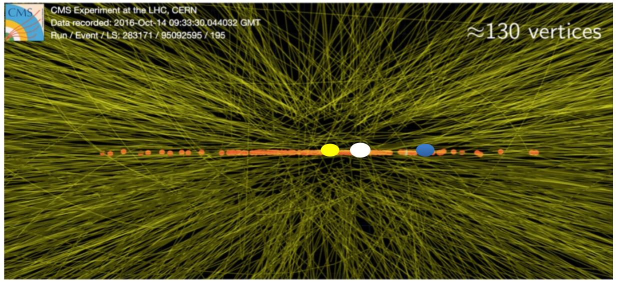
In HEP :

High rate @ high luminosity accelerators;

>140 collision events per bunch crossing at High Luminosity-LHC;

→ Pileup mitigation via TOF requires TOF resolution < 50 ps.

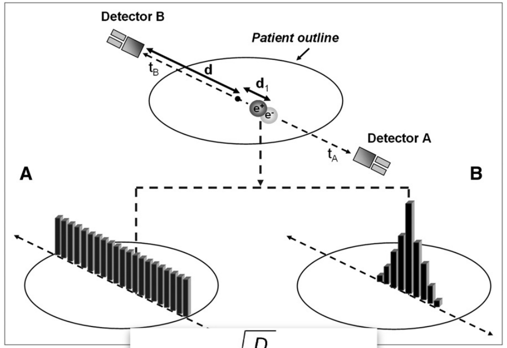
Particle identification



In medical imaging

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

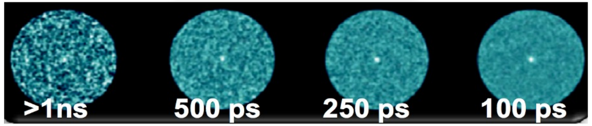


$$SNR_{TOF} \sim \sqrt{\frac{D}{\Delta x}} \cdot SNR_{CONV}$$

In Computed tomograph: TOF CT

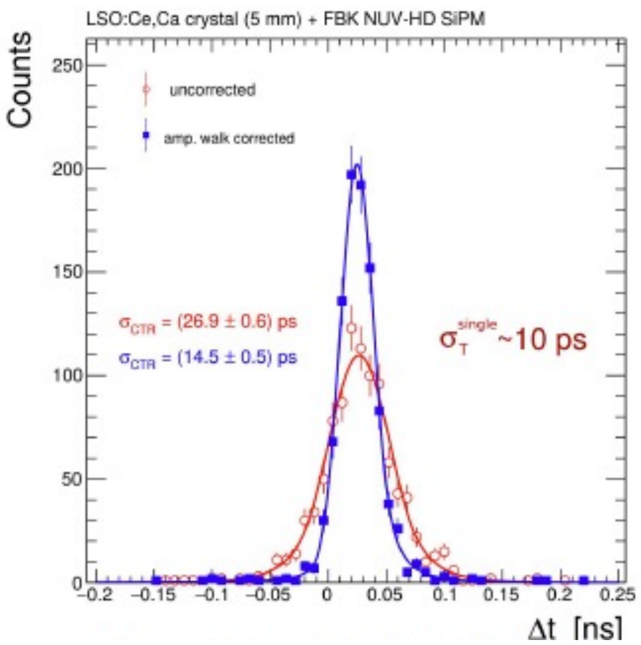
→ Reduce scattered photons contribution

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



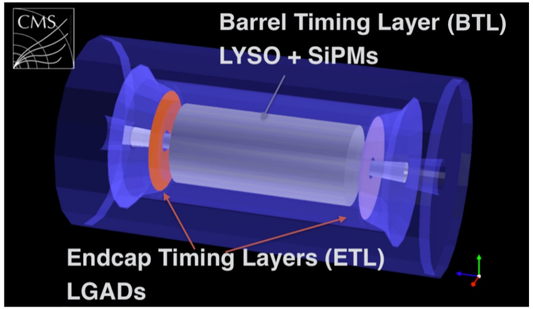
State of the art time resolution with minimun ionising particles (mips)

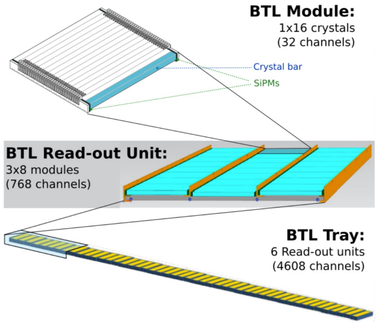
Single LSO:Ce,Ca crystals



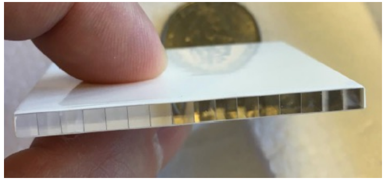
A. Benaglia, et al., NIM A (2016), 830, 30-35

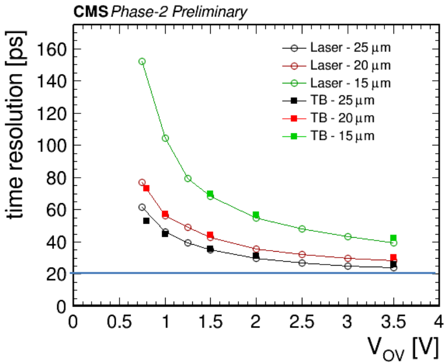
CMS Barrel timing layer for HL-LHC





Fibre array (3*3*57mmm^3)



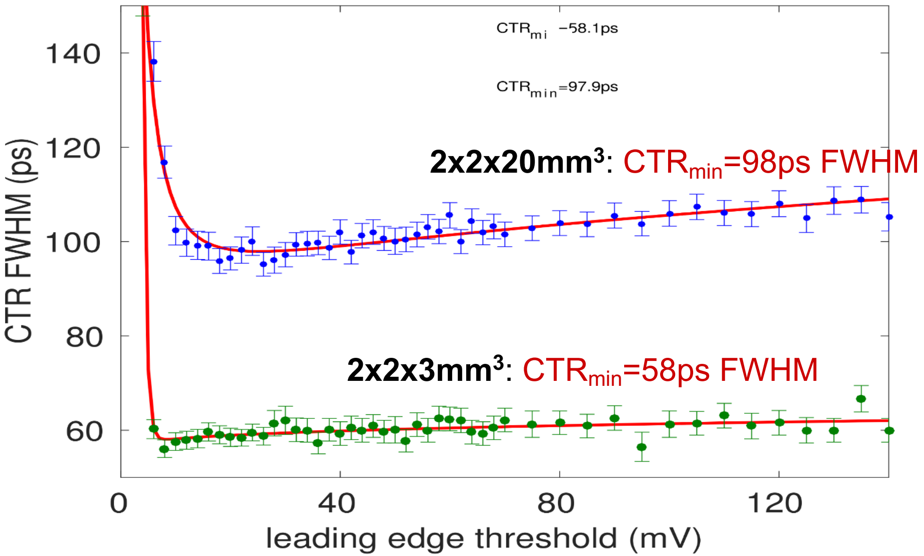


(see talk P. Meridiani Monday)

S. Palluoto, EPS2023

State of the art time resolution with PET size crystal at 511keV

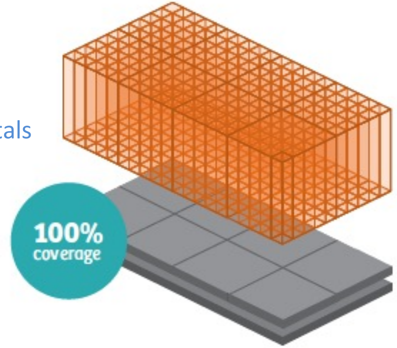
FBK NUV-HD 4x4mm^2, **40x40µm^2 SPAD** + LSO:Ce:Ca



S. Gundacker et al., Phys. Med. Biol. (2019) 64 055012

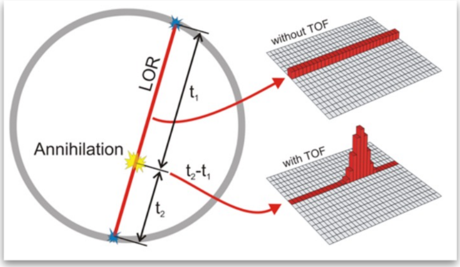
TOF PET SIEMENS: BIOGRAPH VISION

3.2mm section LSO crystals
CTR 215ps

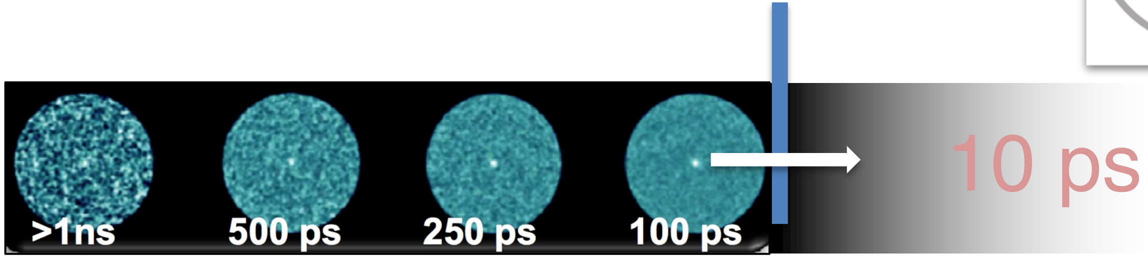


Webpage SIEMENS:, https://static.healthcare.siemens.com/

New challenge in PET
time resolution towards 10ps



https://the10ps-challenge.org



Time resolution	1ns	500ps	250ps	100ps	10ps
Spatial resolution on LOR	15cm	7.5	3.75	1.5cm	**1.5mm**

10ps: Spatial localization directly from TOF (1.5 mm)

Sharp improvement in SNR

Less dose to the patient

More accurate dynamic studies

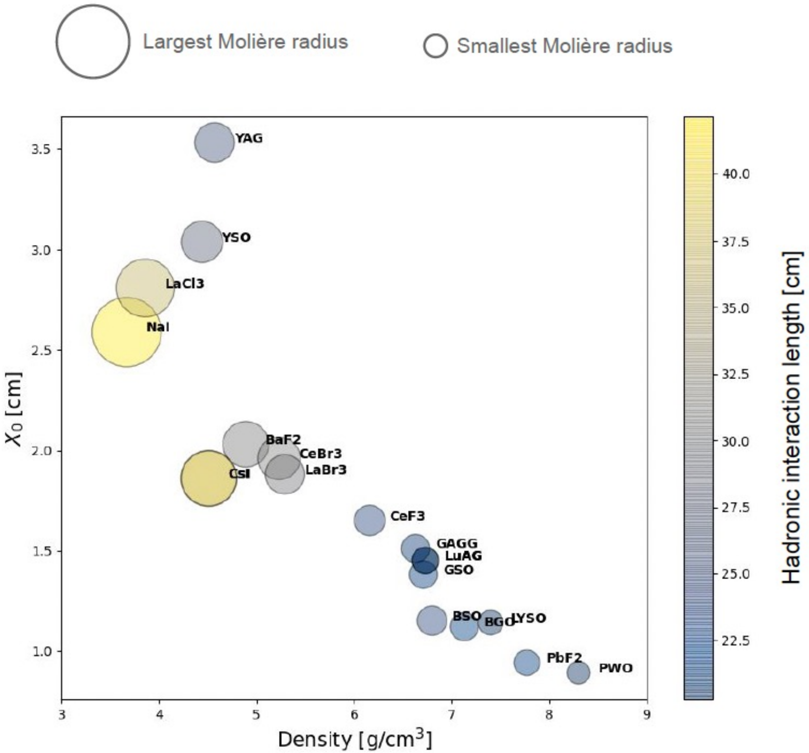
No need for full angular coverage

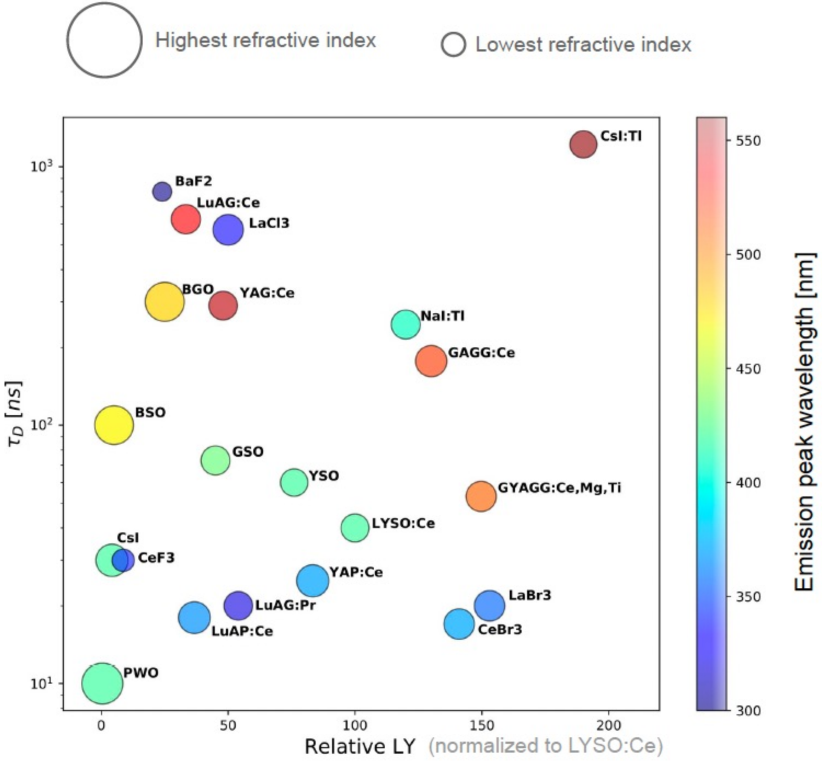
A variety of crystals available

Characteristics of some inorganic crystals

	Na(Tl)	CsI	CsI(Tl)	BGO	PWO	CeF_3	BaF_2	LSO	$LaBr_3$ (Ce)	LuAP Pr/Ce	LuAG: Pr/Ce	GAGG:Ce
ϱ (g/cm^3)	3.67	4.51	4.51	7.13	8.3	6.16	4.89	7.4	5.29	8.34	6.73	6.63
X_o (cm)	2.59	1.86	1.86	1.12	0.89	1.66	2.03	1.14	1.88	1.08	1.41	1.56
Rm (cm)	4.13	3.57	3.57	2.23	2	2.41	3.1	2.07	2.85		2.33	2.1
n	1.85	1.79	1.95	2.15	2.2	1.8	1.5	1.82	1.9	1.97	1.84	1.9
λ (nm)	415	310/420	550	480	420	310	195-220/310	420	356	310/365	290,350/535	520
τ (ns)	230	6/35	10.5	300	10/30	5/30	0.8/630	40	20	20/18	20/70	50-90
LY (ph/MeV)	38000	2000	54000	8000	200	2000	1500/10000	33000	63000	15000/11400	>15000/>25000	>35000

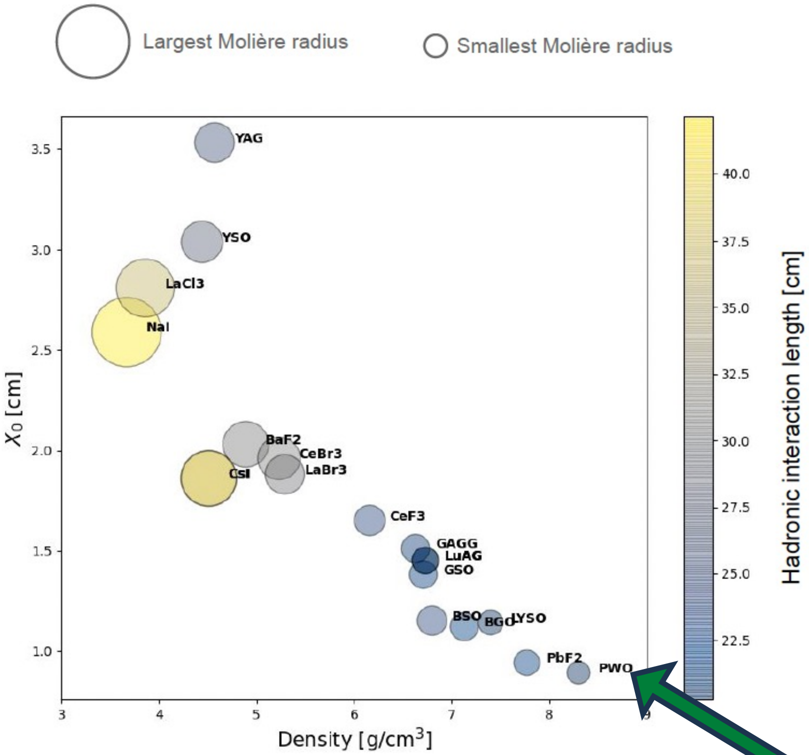
Which one to choose?

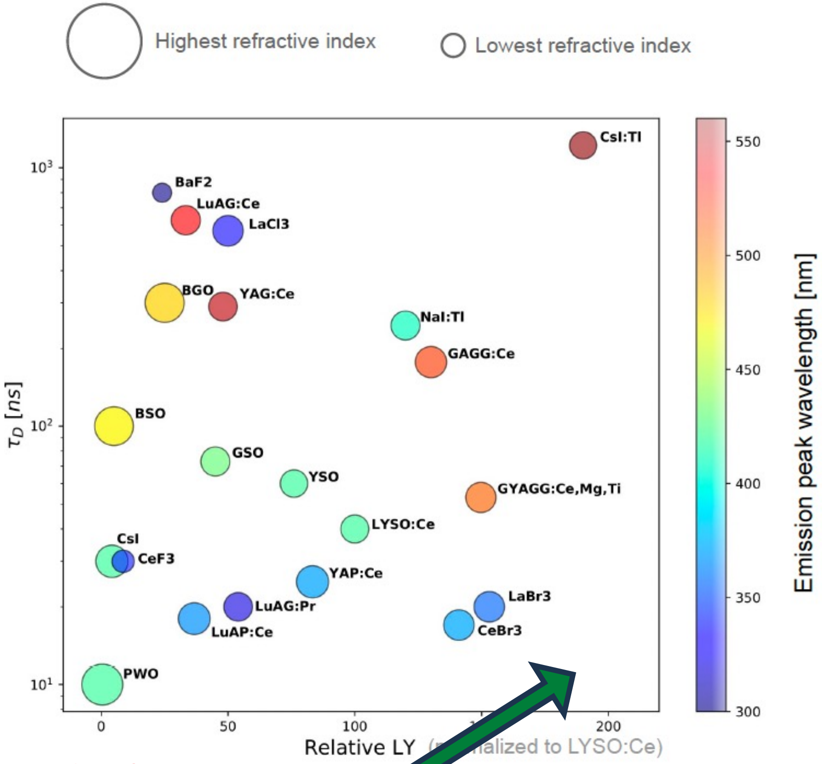




M.T. Lucchini, Scint2022 conference

Which one to choose?





Ideal one?

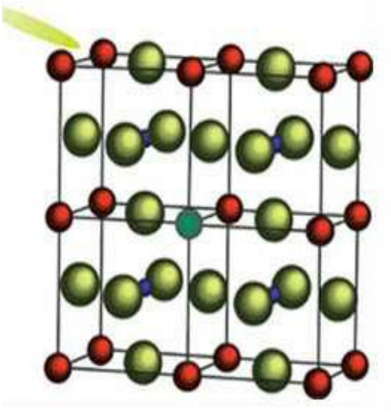
M.T. Lucchini, Scint2022 conference

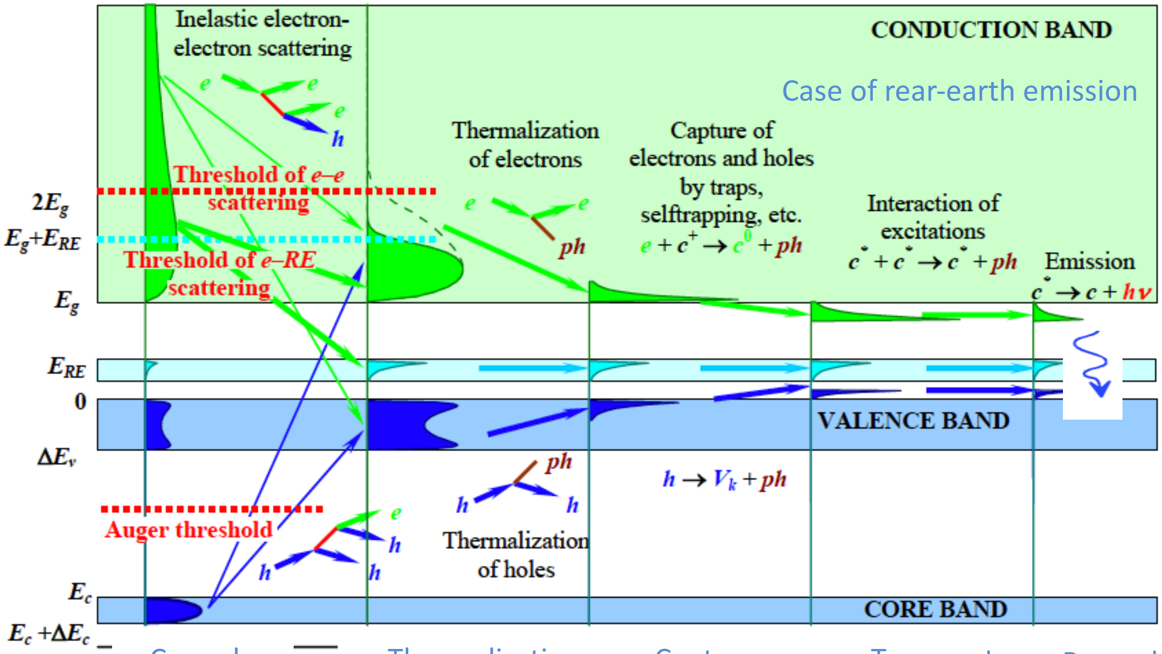
Scintillation: a complex process

From eh pair creation to light emission

A. Vasiliev, SCINT99 conference,

Band structure





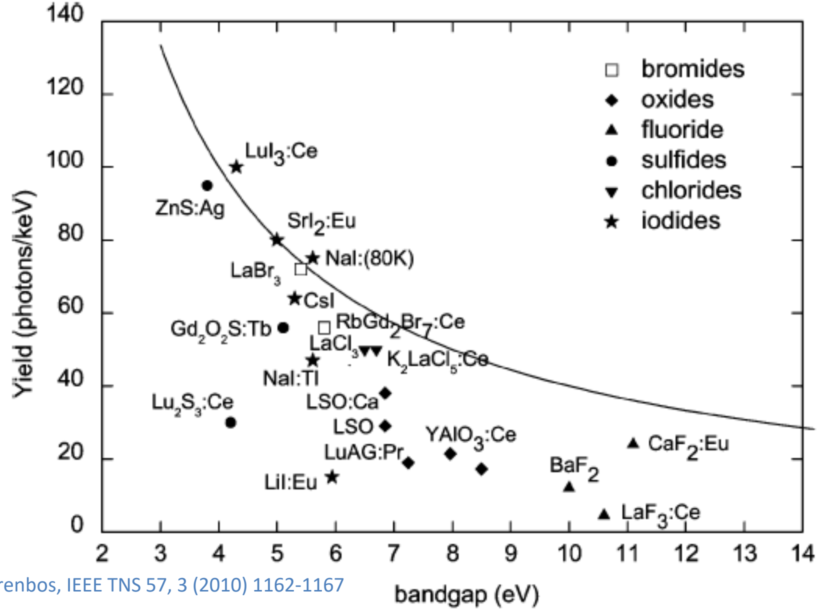
Case of rear-earth emission

Cascade 1fs	Thermalization <1ps	Capture 150 fs- 50 ps	Transport 0-10 ns	Recombination ≳0 .5 ns

Scintillation Yield $N_{eh} = \frac{E_\gamma}{\beta E_g}$ S Q

Scintillation Characteristics

Light Yield

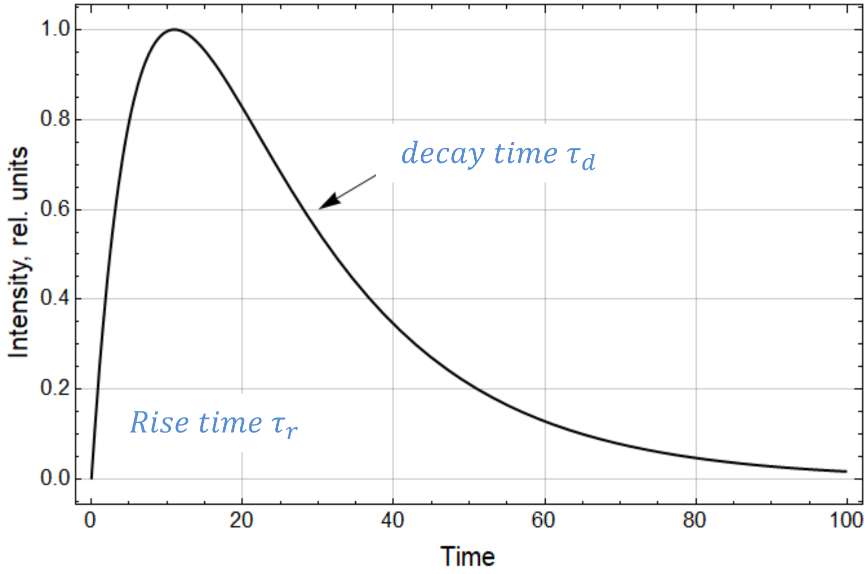


P. Dorenbos, IEEE TNS 57, 3 (2010) 1162-1167

$LY \propto \frac{E_\gamma}{\beta E_g}$ SQ $with\ \beta$ between 2-3

Strong dependence with band gap energy

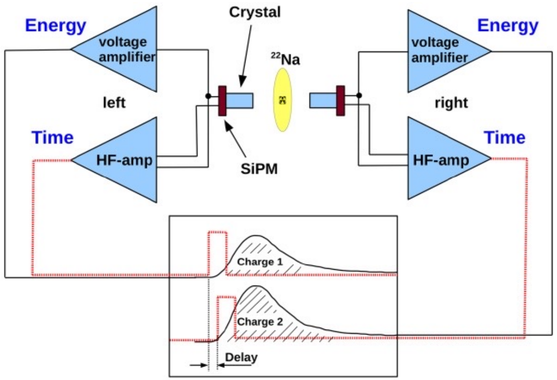
Pulse shape spectra

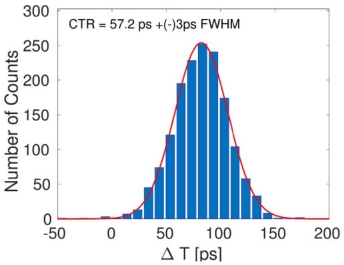


$$f(t|\theta) = \sum_{i=1}^{3} R_i \cdot \frac{\exp\left(-\frac{t-\theta}{\tau_{d,i}}\right) - \exp\left(-\frac{t-\theta}{\tau_r}\right)}{\tau_{d,i} - \tau_r} \cdot \Theta(t-\theta)$$

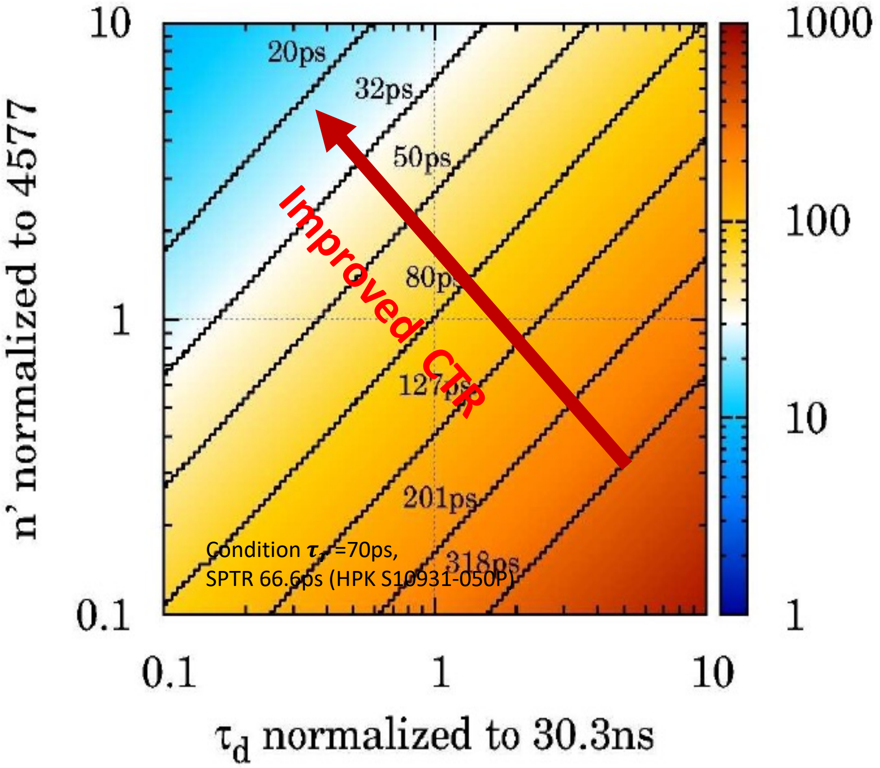
Scintillation time characteristics

Coincidence time resolution (CTR)





$$CTR \ \alpha \sqrt{\frac{\tau_d \tau_r}{Light\ Output}}$$



S. Gundacker et al., Phys. Med. Biol. (2019) 64 055012

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

Various emission process

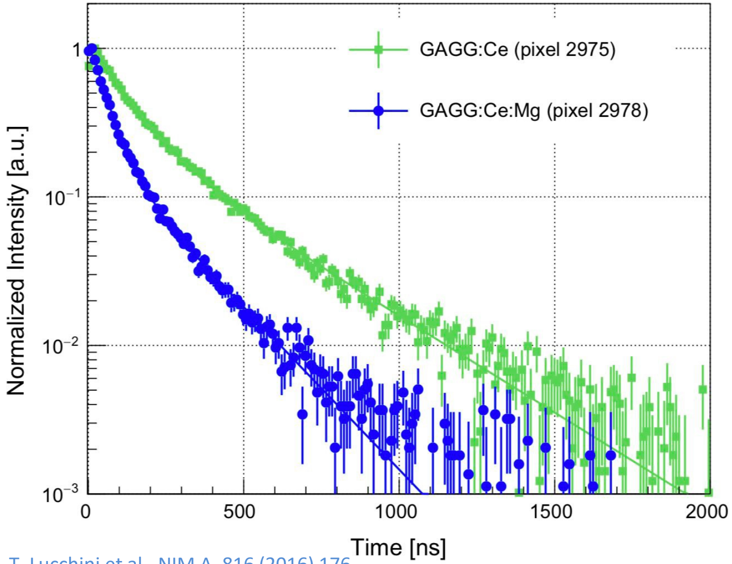
- Excitonic emission (STE, excitations of anion complexes)
- Emission of activators (Ce, Pr, …) Codoping:
- **Cherenkov radiation**
- **Crossluminescence**
- **Hot intraband luminescence (HIL)**
- **Quantum confinement driven luminescence:**

Slow

↓

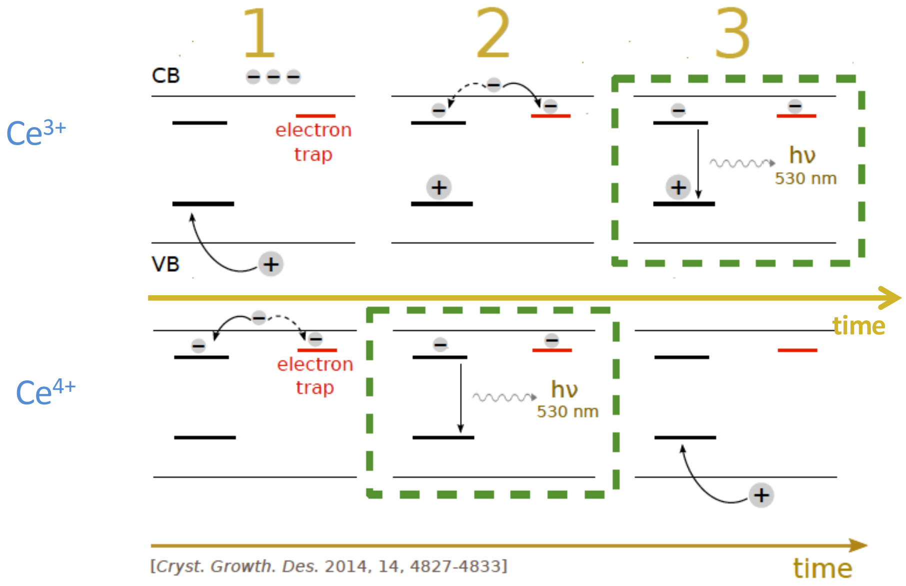
Ultra fast

Scintillator engineering example: codoping Ce, Mg in garnet



M. T. Lucchini et al., NIM A, 816 (2016) 176

Faster decay time with codoping Ce^{3+}/Mg^{2+}



Mg^{2+} increase Ce^{4+} centers which can directly compete with any electron trap for electron capture in the first instants of scintillator mechanism

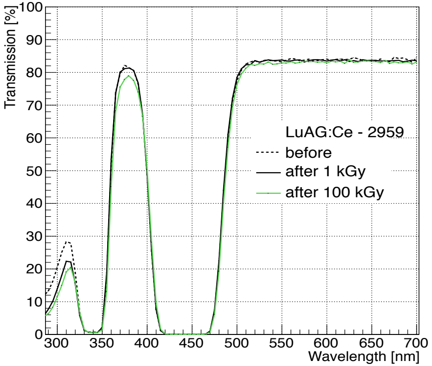
=> Expected faster decay time and lower slow component

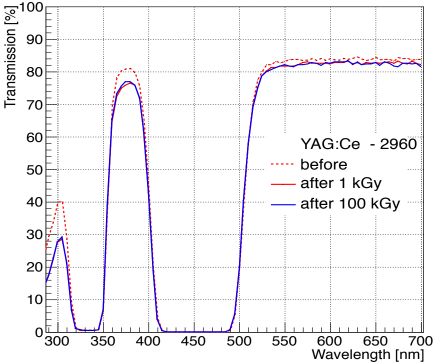
M. Nikl, A. Yoshikawa, Adv. Optical Mater. 2015, 3, 463–481
M. Nikl et al. Cryst. Growth Des. 2014 , 14 , 4827 .

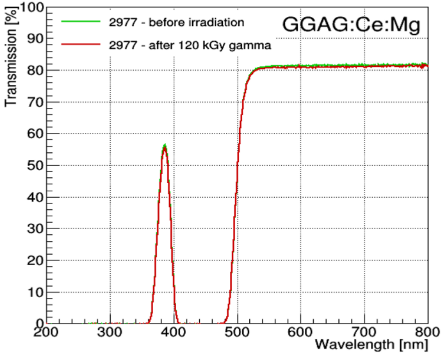
Radiation hardness of garnet scintillators

LuAG | YAG | GAGG

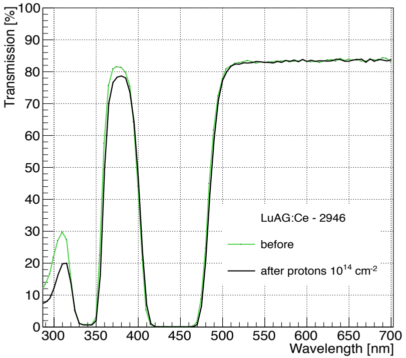
Gamma

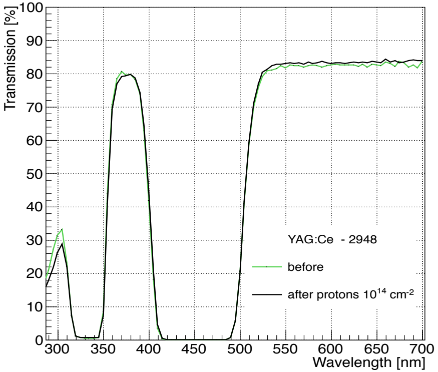


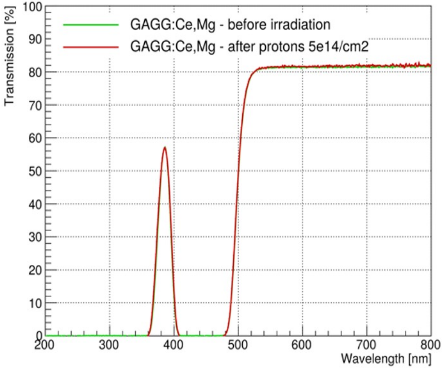




protons





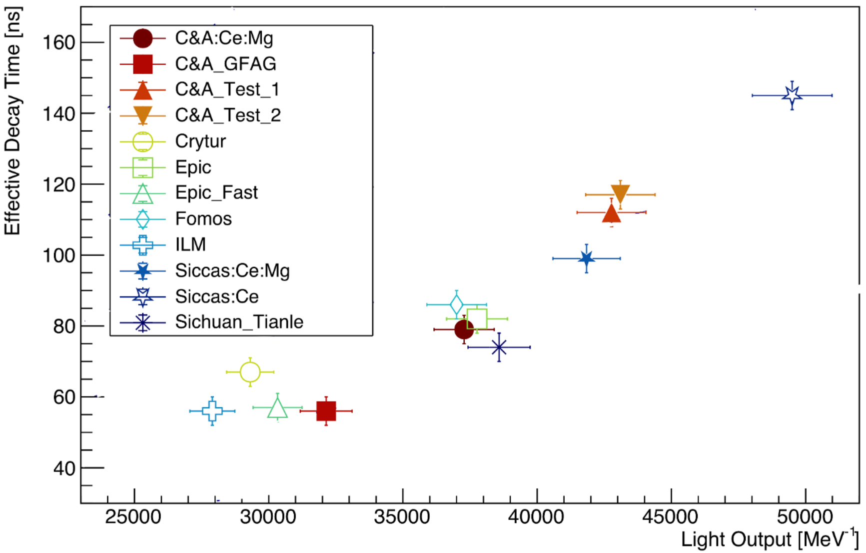


M. T. Lucchini, et al.,, IEEE Transactions on Nuclear Science (2016), 63, 2
E. Auffray, et al, Rad. Phys.Chem. (2019), 164, 108365
V. Alenkov, et a., NIM A (2019), 916, 418 226{229

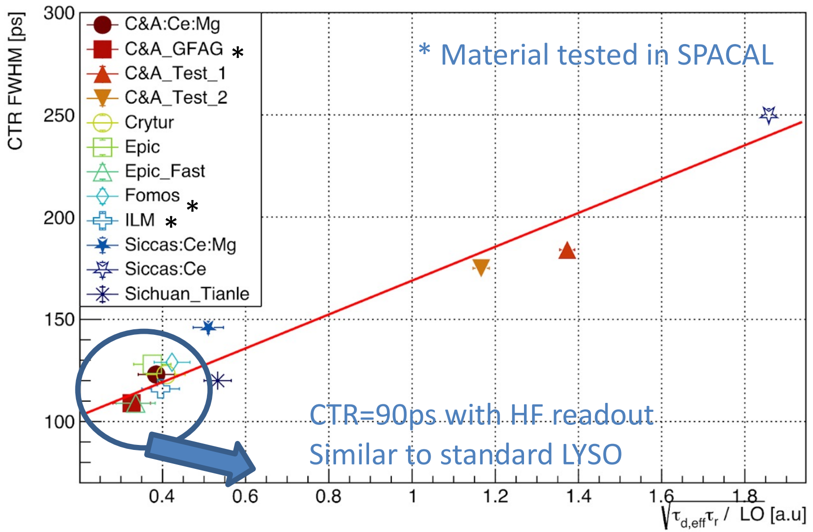
Very Good radiation tolerance under gamma & proton radiations

GAGG ($Gd_3Al_2Ga_3O_{12}$): Tunable properties with composition

Effective decay time versus Light output



Coincidence time resolution (CTR) versus photon density



Correlation between Light output and decay time

CTR inversely proportional to the photon time-density ($\frac{LO}{t_{d\,eff}}$):

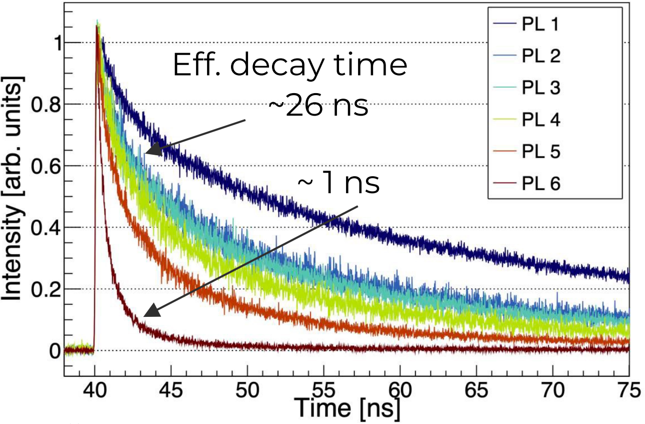
Candidate for central part of LHCb phase II calorimeter (see talk P. Roloff Monday)

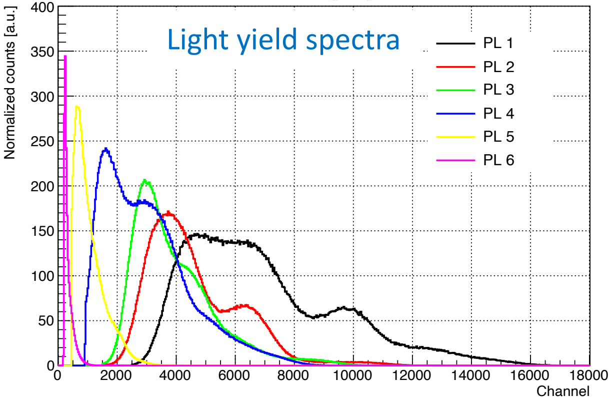
L. Martinazzoli et al., NIM A, 2021, 165231

Further acceleration of the emission

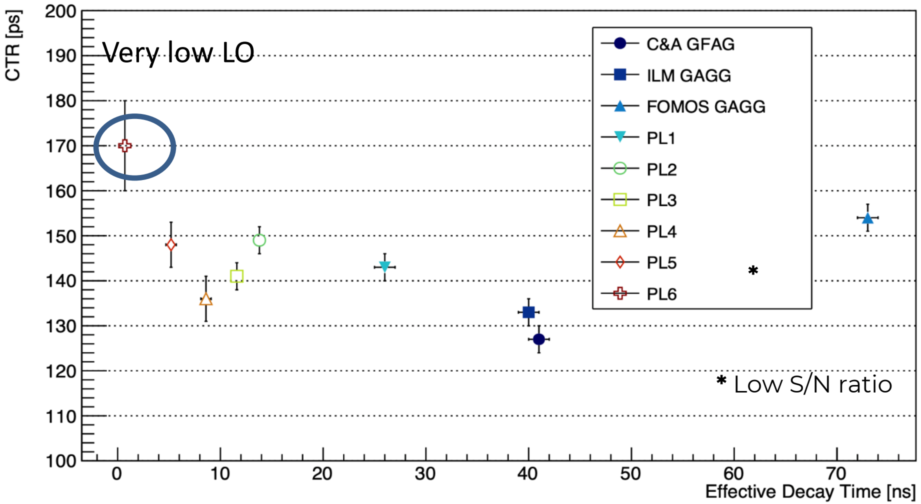
Heavy codoping Ce^{3+}/Mg^{2+}

Scintillation decay - Pulsed X-Rays





Coincidence time resolution vs effective decay time

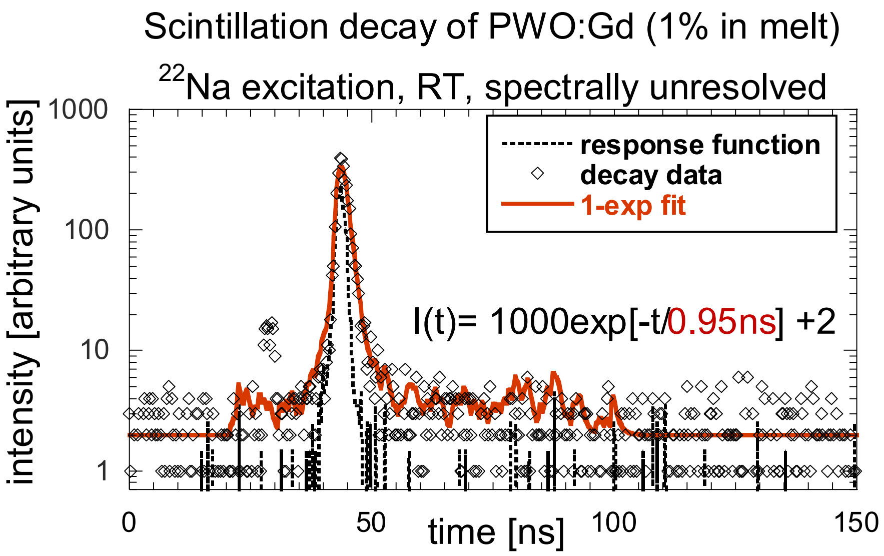


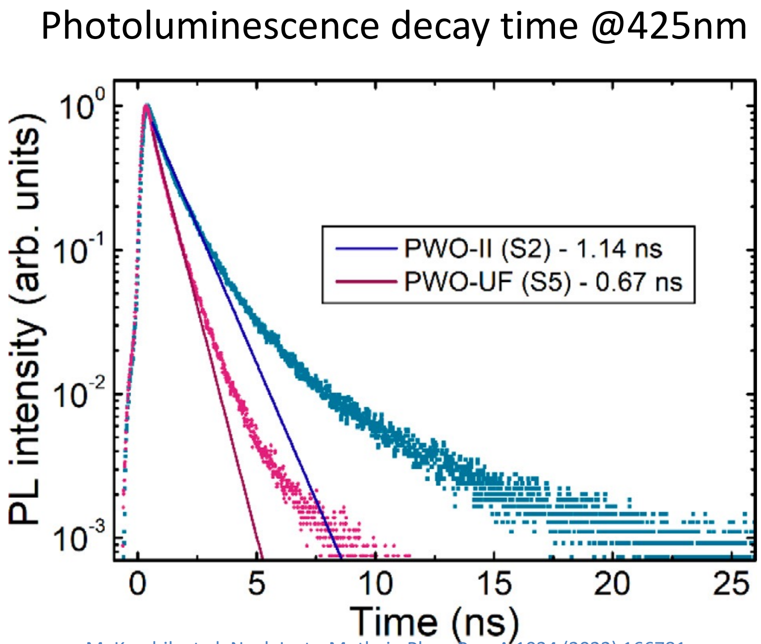
No major loss of time resolution!
Decay time decrease compensated the Light output reduction
=> the same photon time-density
R&D on production on going

L. Martinazzoli et al., Mater. Adv., 2022, 3, 6842

Towards very fast PWO

Many developments on PWO to decrease decay time toward sub ns domain with heavy doping:





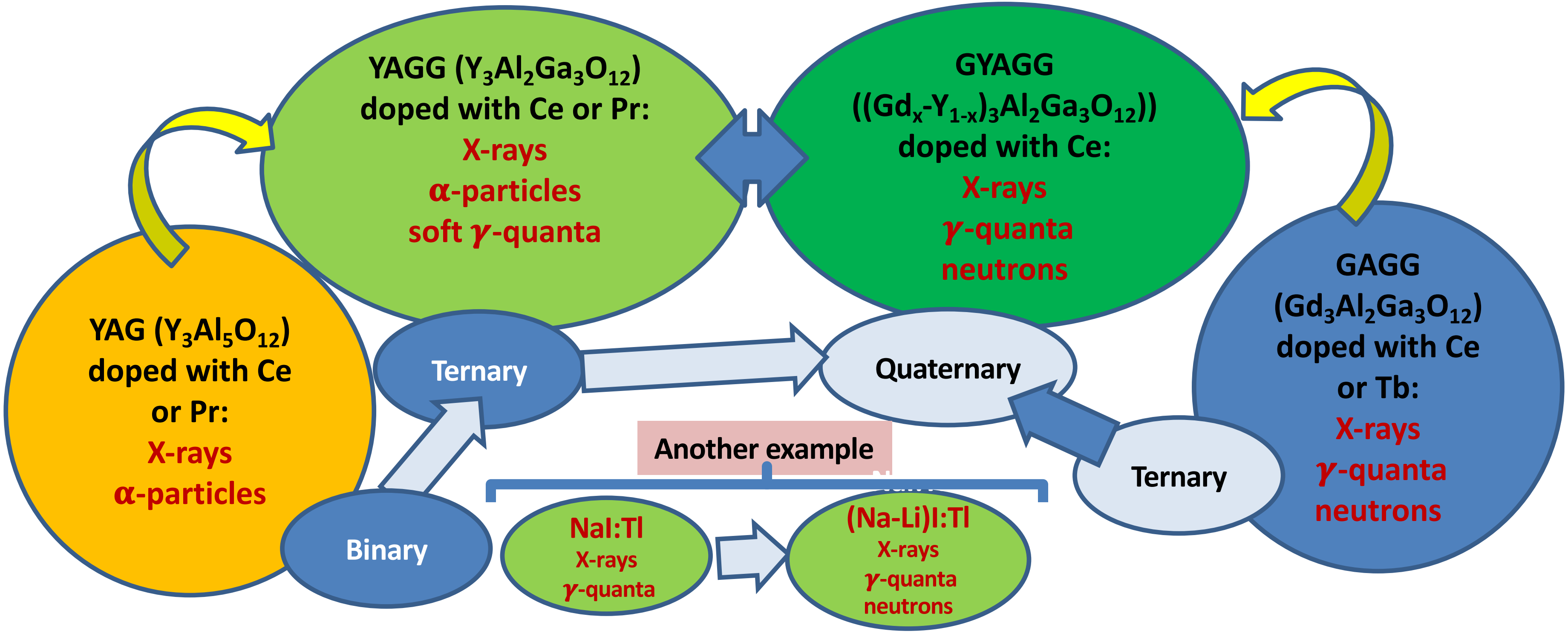
M. Korzhik et al, Nucl. Instr. Meth. in Phys. Res. A 1034 (2022) 166781

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)

Candidate for KLEVER &CRILIN calorimeter

Mixed materials: concept of multipurpose scintillation materials

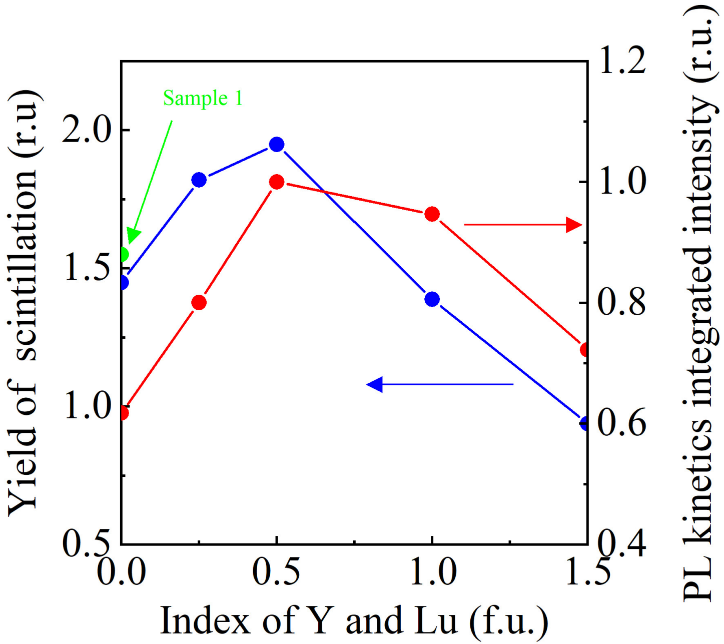
Possibility to modify crystal composition

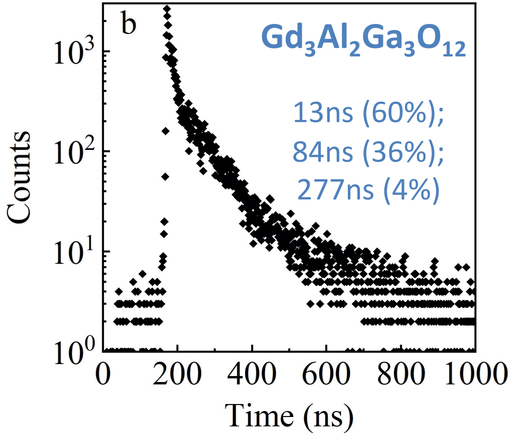


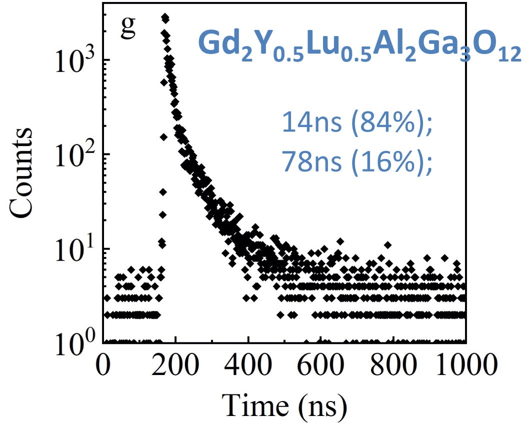
Courtesy M. Korzhik, RINP, Minsk

$(Gd,Y,Lu)_3Al_2Ga_3O_{12}$:Ce, Mg

Light Yield and photoluminescence

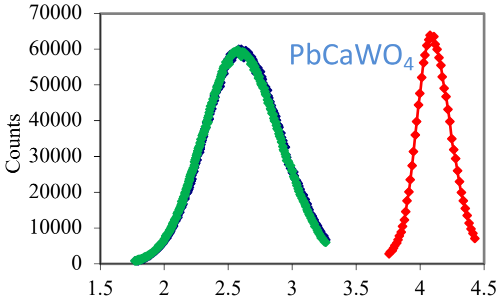


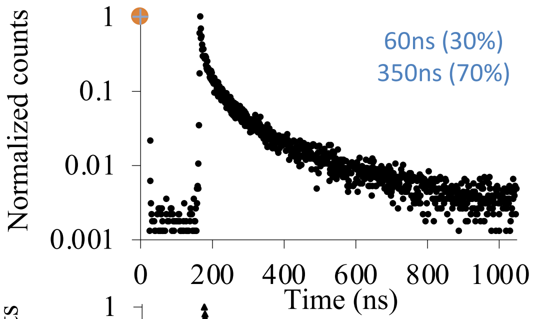


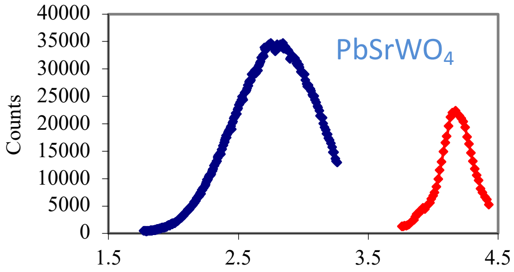


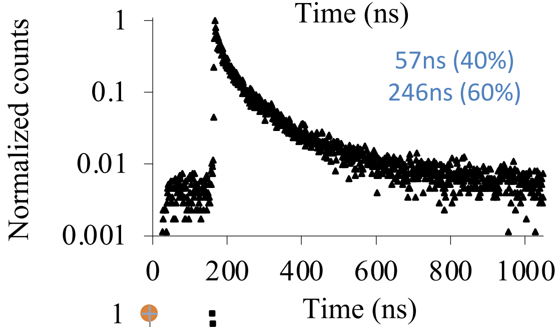
D. Kuznetsova et al., Journal of Applied Physics, 2022; 132 (20): 203104.

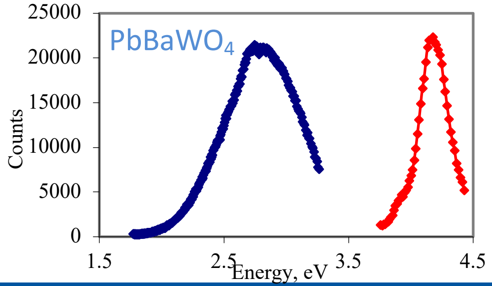
New mixed tungstate $(Pb,Ca,Sr,Ba)WO_4$

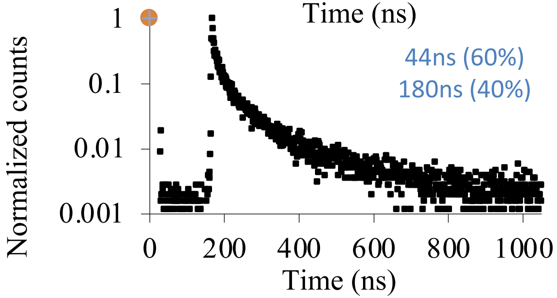












Courtesy M. Korzhik, RINP, Minsk

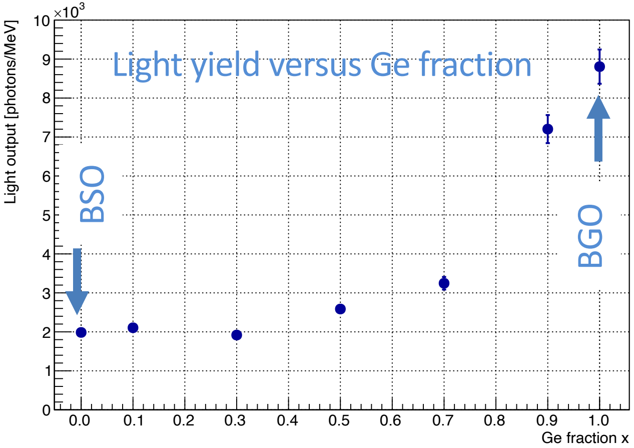
Mixed tungstate properties

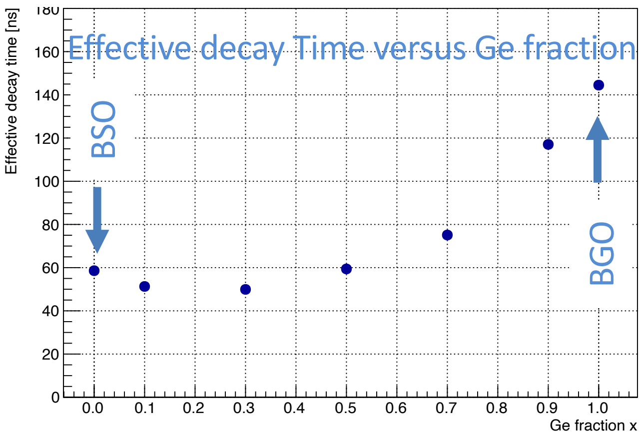
Compound	$PbWO_4$	$CaWO_4$	$SrWO_4$	$BaWO_4$	$(Pb, Ca)WO_4$ (PCWO)	$(Pb, Sr)WO_4$ (PSWO)	$(Pb, Ba)WO_4$ (PBWO)
Density, g/cm^3	8.28	6.12	6.03	6.38	7.20	7.15	7.33
Effective charge, Z_{eff}	76	66	64	65	72	71	71
Photo-absorption coefficient at 511 keV, cm^{-1}	0.485	0.222	0.197	0.223	0.359	0.340	0.350
Radiation length Xo, cm	0.89	1.49	1.50	1.33	1.11	1.11	1.07
Moliere radius R_M, cm	1.91	2.28	2.40	2.36	2.09	2.12	2.11
LY, ph/MeV (γ-quanta)	200	14400	1200	>100	7000	8700	5500
Parameters of the scintillation kinetics, ns (%)	1.8(60) 6(40)	8200	522	>10	60(30) 350(70)	57(40) 246(60)	44(60) 180(40)
*Effective decay constant, ns					263	170	90

Courtesy M. Korzhik, RINP, Minsk

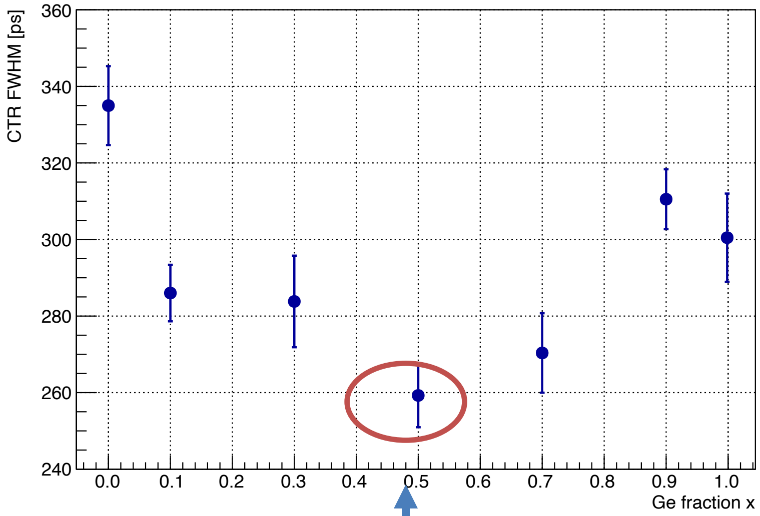
Mixed Material: BGO-BSO ($Bi_4(Ge_xSi_{1-x})_3O_{12}$)

To tune the material properties





Coincidence time resolution @511Kev versus Ge fraction



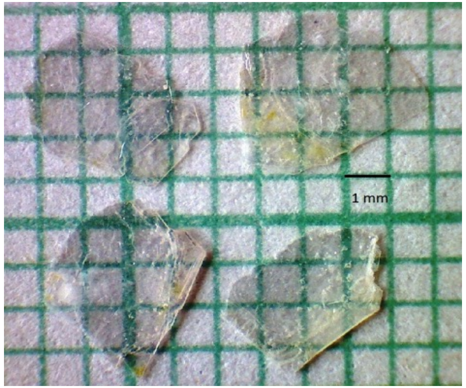
BSO BGO

Optimal Ge fraction for time resolution

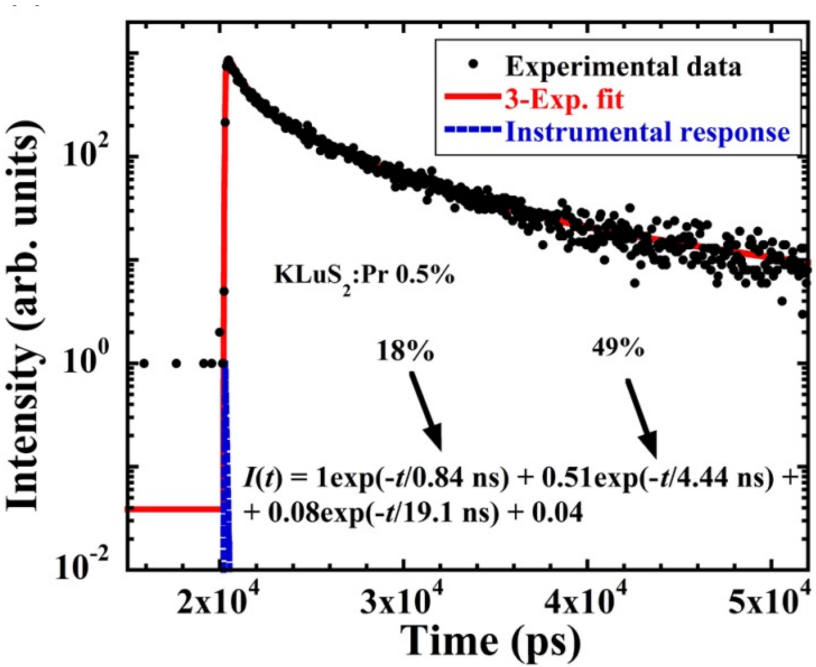
R&D on production on going

R. Cala et al, NIMA, A 1032 (2022) 166527

$KLuS_2:Pr^{3+}$ for fast timing



Light Yield: 7000ph/MeV



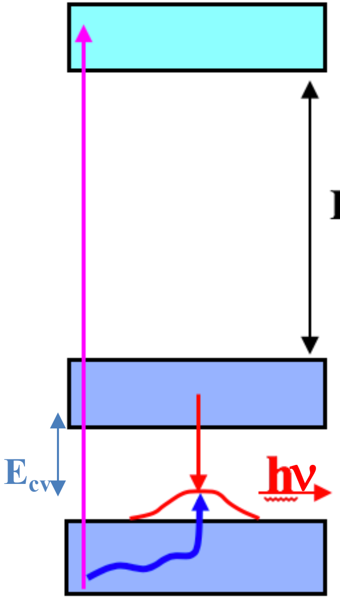
=> 1.6× – 2.5× more scintillation light in first nanosecond compared to LYSO:Ce,Ca
=> potentially better time resolution

FZU Fyzikální ústav Akademie věd České republiky

Jarý et al. Phys. Rev. Applied 19, 034092 (2023))

Crossluminescence material

Radiative transition between the core- and valence bands.



$E_{cv} < E_g$

Many possible materials

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

Compilation of CL data at 293 K

	E(C − V) (eV)	E(G) (eV)	Theoretical	Observed (eV)	λ (nm)	Light yield (photons/MeV)	τ (ns)	Density (g/cm³)	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 UV emission

Crossluminescence material: BaF_2

Compilation of CL data at 293 K

	$E(C-V)$ (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]
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RbBr	6.7–9.5	7.4	/						
RbI	5–10	6.1	/	STE					
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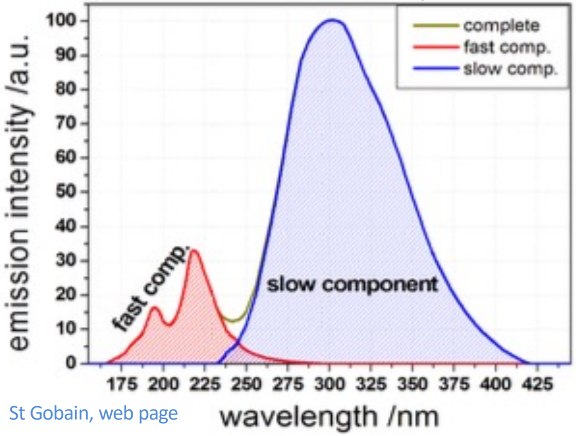
CsCl	1–5	8.3	+	4–5.5	240, 270	900	0.9	4.0	[6, 14, 15, 17, 18]
CsBr	4–6	7.3	+	4.5–6.5	250	20	0.07	4.4	[6, 14, 15, 18]
CsI	0–7	6.2	/	–/STE					
CaF_2	12.5–17.3	12.6	–	–/STE					[1]
SrF_2	8.4–12.8	11.1	/	–/STE					[1]
BaF_2	4.4–7.8	10.5	+	5–7	195, 220	1400	0.8	4.9	[1, 3, 4, 9]
$K_xRb_{1-x}F$				5–6/8					[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]

BaF_2 emisison spectra



St Gobain, web page

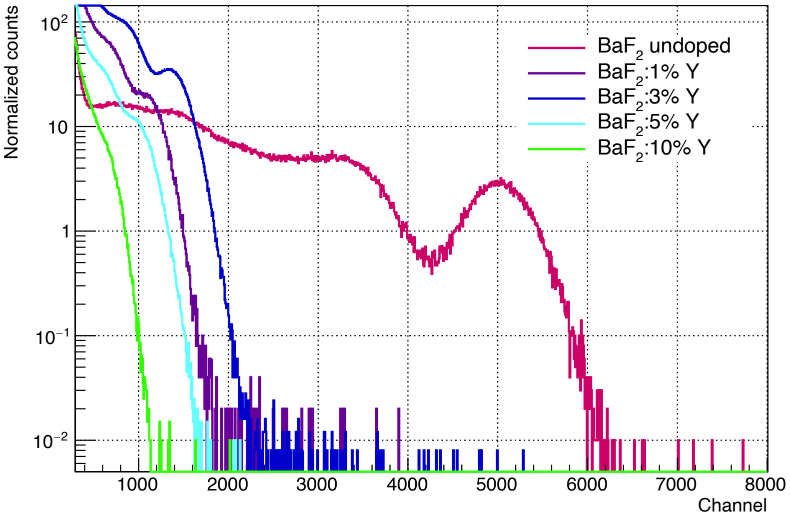
BaF2 was proposed in 90's for ECAL by L* Collaboration, Letter of Intent to the SSC Laboratory

Sub ns emission but in UV & additional slow component

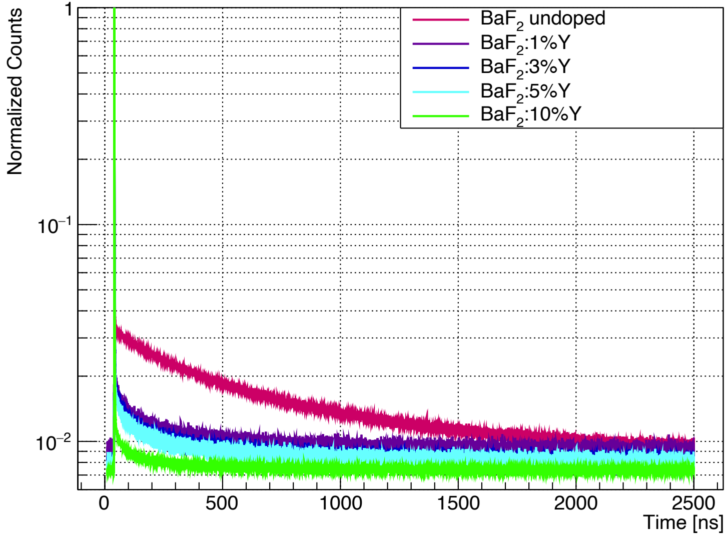
https://lss.fnal.gov/archive/other/ssc/sscl-sr-1154.pdf
R. Zhu, NIMA A 340 (1994) 442-457

Suppression of slow component in BaF_2 with Y codoping

LY spectra



Decay time spectra



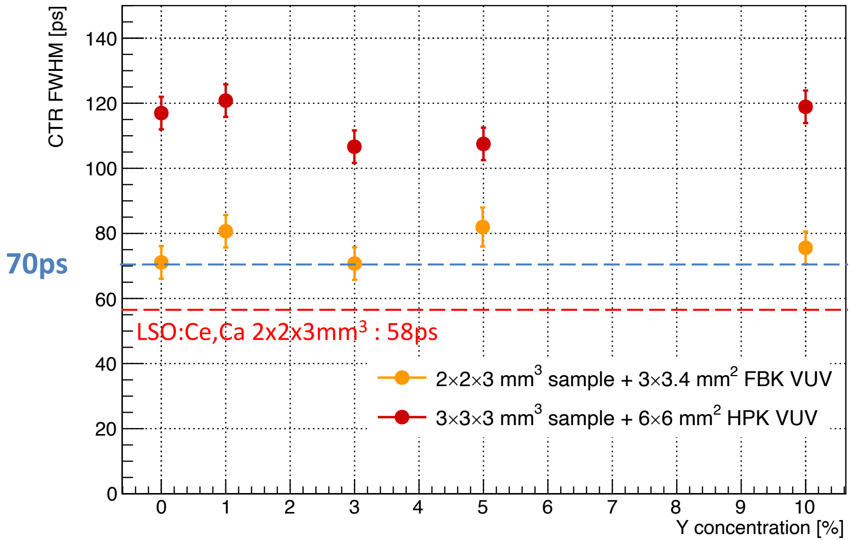
With Y doping:
No change of fast decay decay time, only slow component decrease

R. cala' et al, paper under preparation

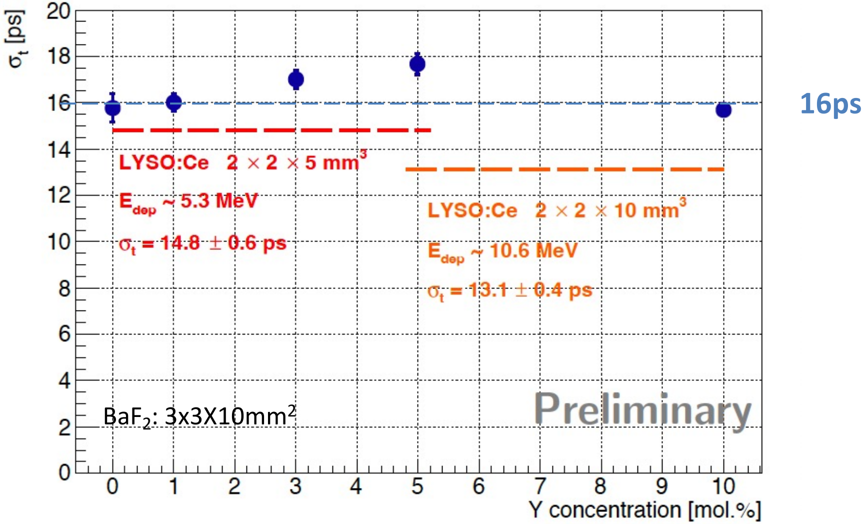
See :
J. Chen, et al., IEEE Trans. Nucl. Sci., vol. 65, no. 8, pp. 2147-2151, 2018.
S. Gundacker et al., Phys. Med. Biol. 66 (2021) 114002

Suppression of slow component in BaF_2 with Y codoping

CTR at 511keV



Time resolution with mips

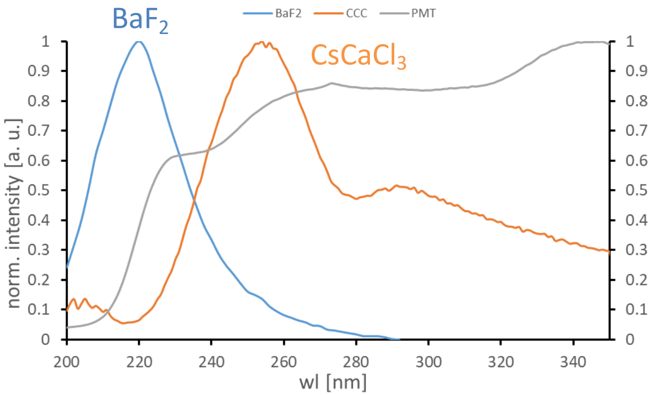


Similar time resolution than LS0 but SiPM with lower PDE without optical coupling

R. Cala, et al. CERN, paper in preparation

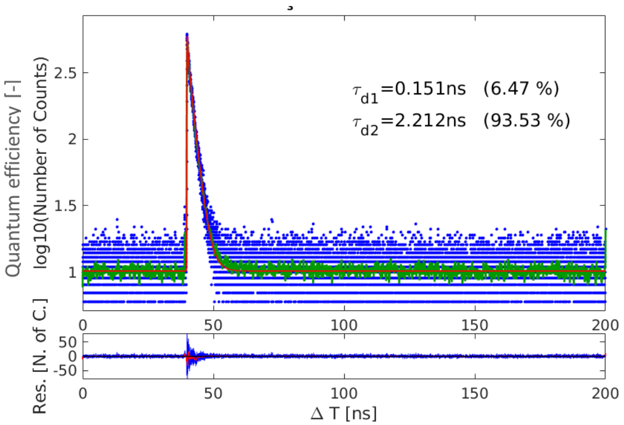
Development of cross luminescence material more in UV/visible region

Emission spectra

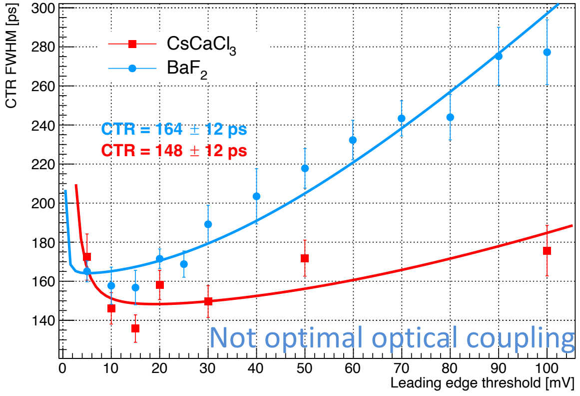


Courtesy V. Vanecek, M. Nikl, FZU Prague
Data for BaF_2 from M. Laval et al. , NIM Phys. Res., 206 (1983) 169–176

Decay spectra



CTR @ 511keV



$CsCaCl_3$: 2 emissions @ 260nm & 290nm
2 fast decay times: 0.15ns, 2.2ns
Same CTR than BaF_2

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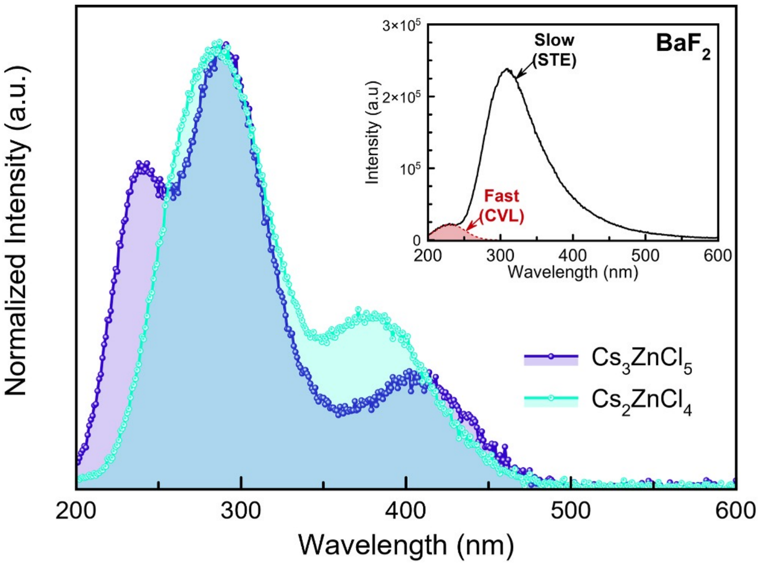
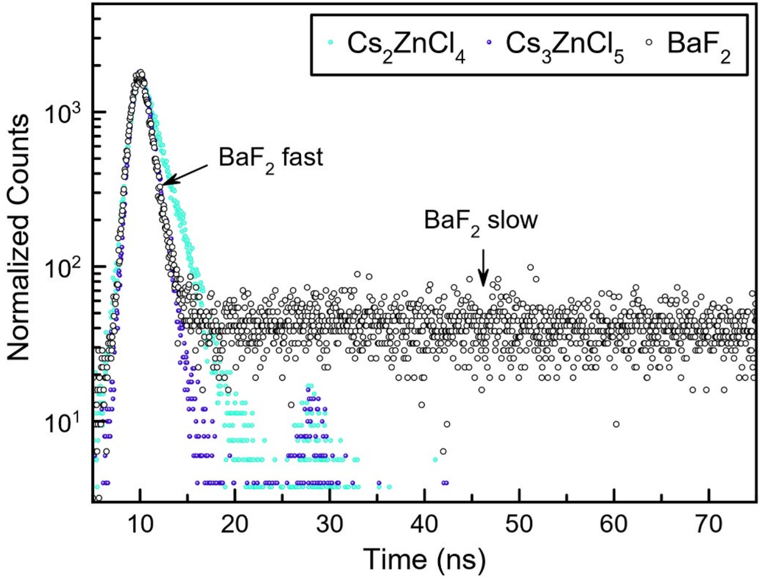
V. Vanecek et al., Optical Materials X 12 (2021) 100103

Development of cross luminescence material more in UV/visible region





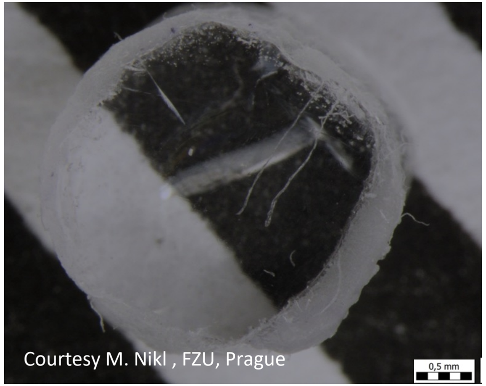
$CsZnCl_{4,5}$: emissions @ 300nm & 370nm
fast decay times and no slow components

D. Rutstrom et al, Optical Materials 133 (2022) 112912

Further development of cross-luminescence materials ongoing in different labs

- Collaboration between FZU, Prague and IMR, Tohoku University, Sendai to growth fluoride compounds by micro-pulling-down method aiming:
  - Better spectral matching with detector
  - Air stable
  - Incorporation of heavy elements

  First attempts:
  - $CsSrF_3$, $CsCaF_3$ → high evaporation of CsF
  - $CsMgF_3$ → unstable phase
  - $Cs_4Mg_3F_{10}$ → stable, non-hygroscopic

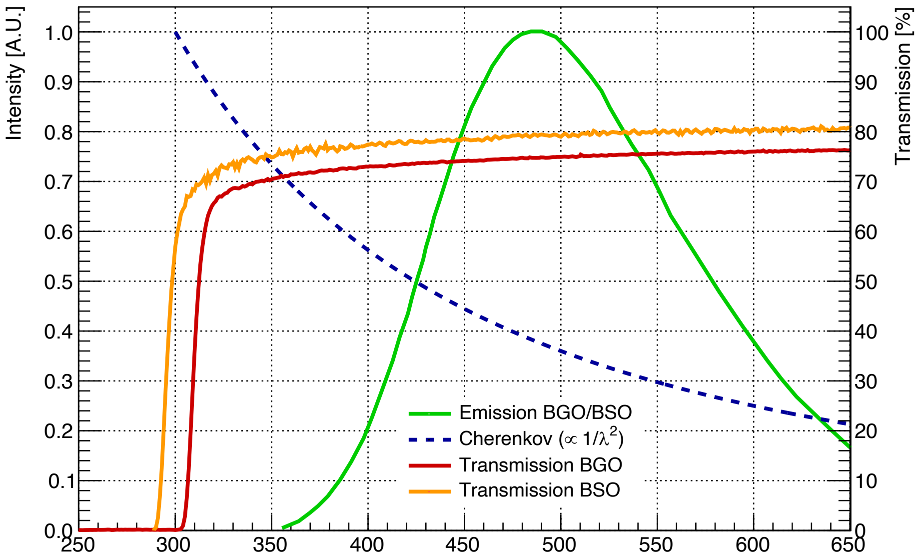


Polished sample prepared from mPD grown $Cs_4Mg_3F_{10}$ crystal

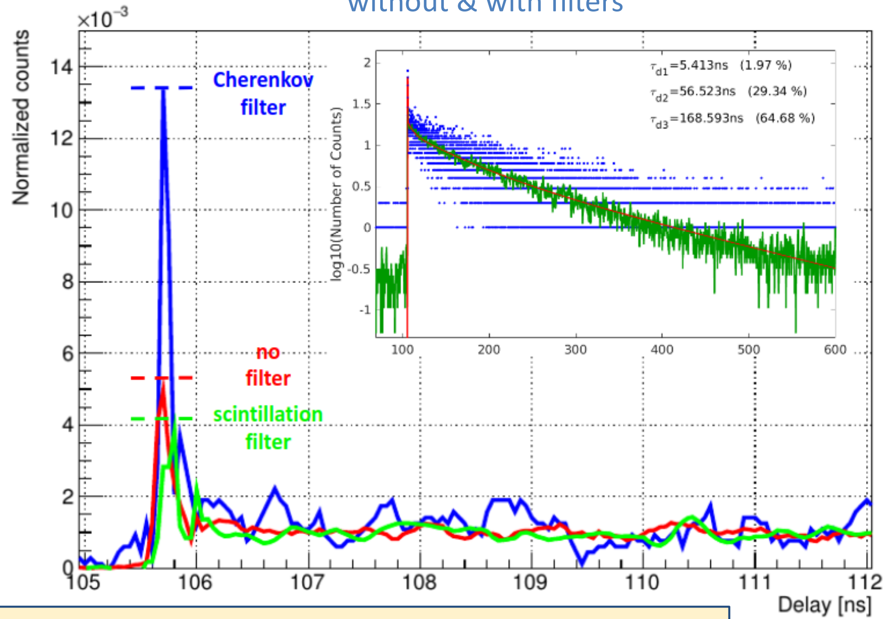
- Group of Tartu, Estonia:
  - Exploring ternary fluorides like K_2GeF_6: ultra fast emission

Exploitation of Cherenkov/scintillation in intrinsic scintillating crystals

BGO and BSO transmission spectra



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



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

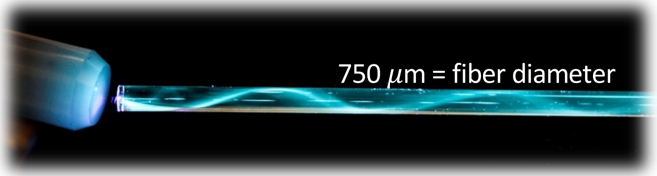
BSO (or mixed BGSO) is faster than BGO and has higher LY than PWO

⇒ Promising candidate for dual readout homogenous calorimeter

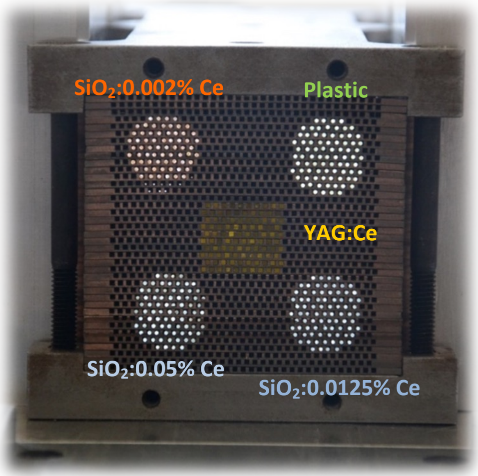
R. Cala et al, NIMA 1032, 2022, 166527

Exploitation of Cherenkov/scintillation in Silica doped fibres

SiO2:Ce fibres Milano/Polymicro

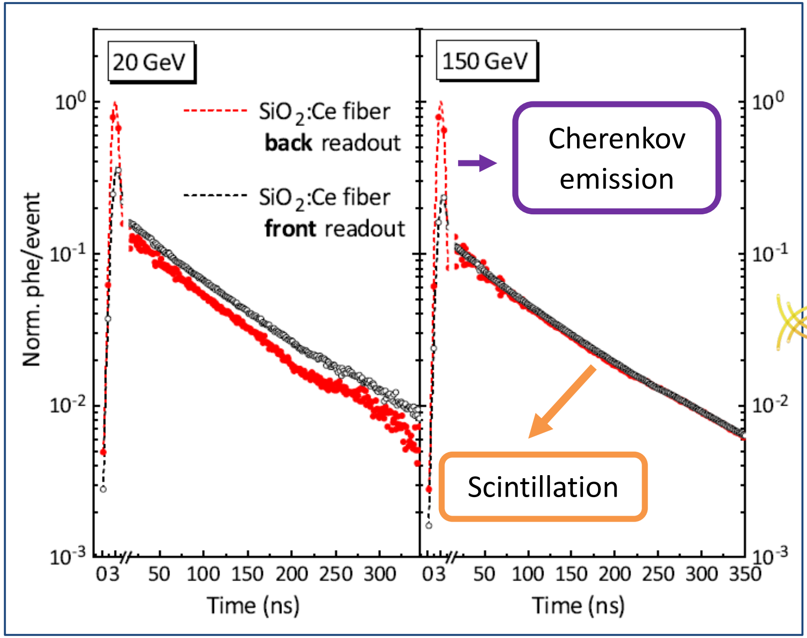


Test with 20GeV in CERN SPS



F. Cova et al., Phys. Rev. Appl. 11 (2), 024036 (2019)

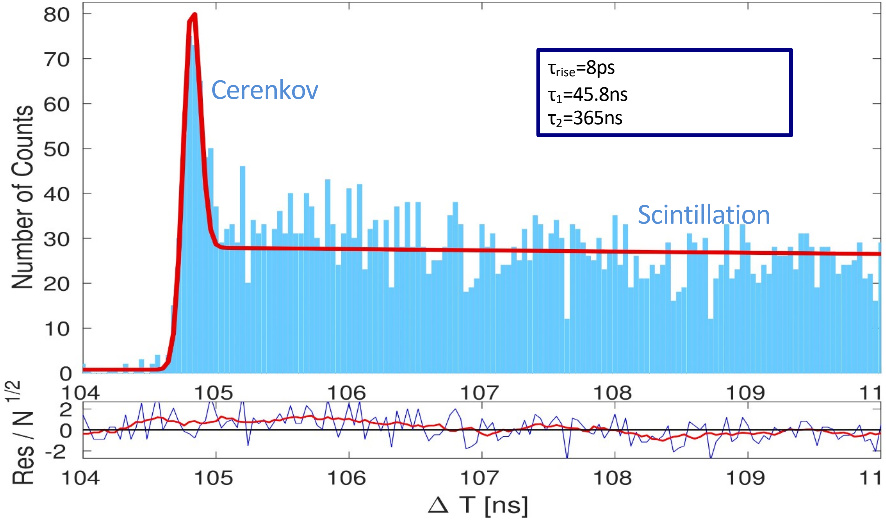
Dual read-out of Cherenkov and scintillation light simultaneously with the same SiO_2:Ce fibre



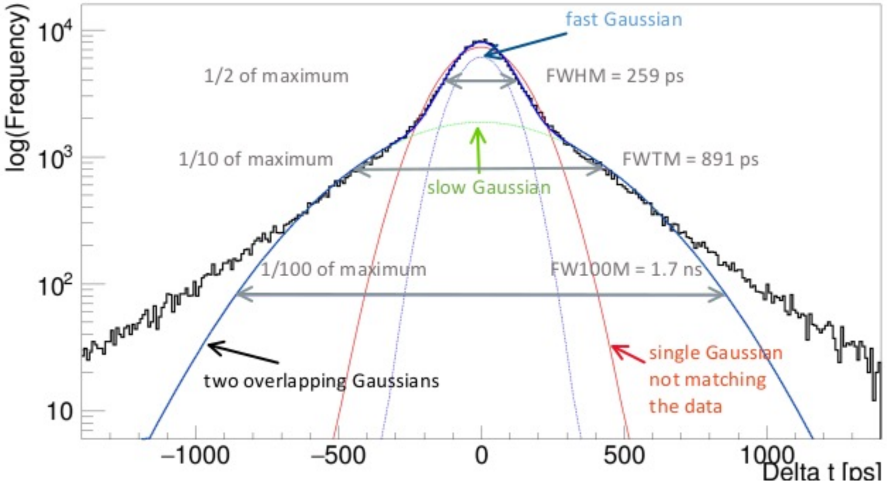
Cherenkov exploitation to improve time resolution

Example of BGO

Decay time spectra with 511KeV



Coincidence time resolution @ 511KeV



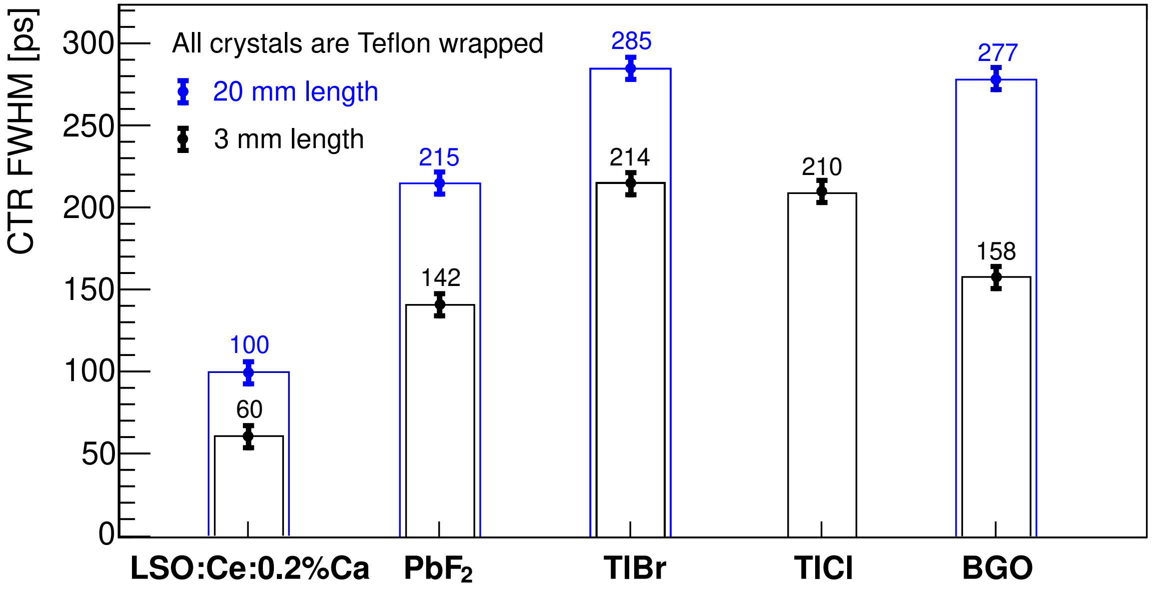
S. Gundacker et al. (2019) Phys. Med. Biol. 64 055012

N. Kratochwil et al (2020), Phys. Med. Biol. 65 115004

N Kratochwil et al (2020) IEEE TRPMS 2020.3030483

Cherenkov exploitation to improve time resolution

CTR at 511KeV

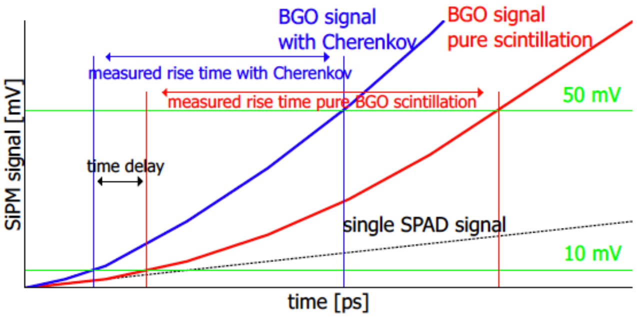


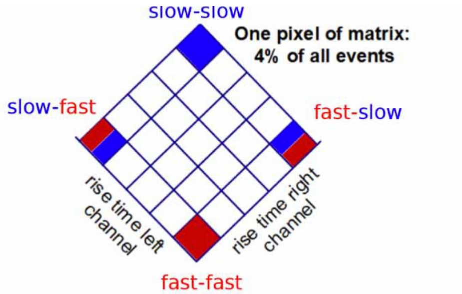
S. Gundacker et al, Phys. Med. Biol. 65 (2020) 025001 (LSO& BGO)
N. Kratochwil et al 2021 Phys. Med. Biol. 66 195001 (PbF_2)
G. Terragni et al., Front. Phys. 2022 10:785627., (TlCl& TlBr)

PbF2: candidate for Klever & CRILIN calorimeter

Cherenkov exploitation to improve time resolution
Further Improvement

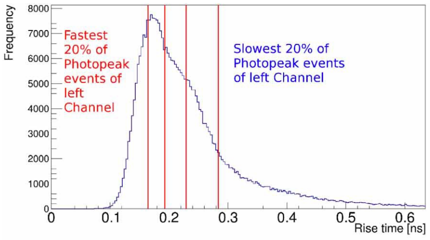
Variation of rise time with Cherenkov events

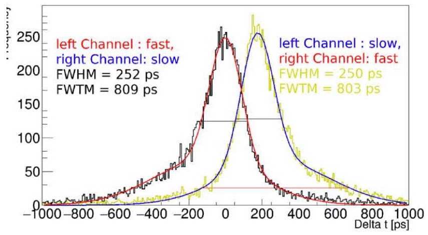


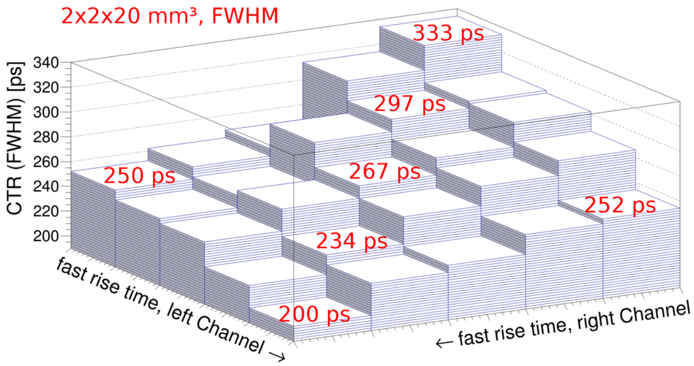


N. Kratochwil et al (2020), Phys. Med. Biol. 65 115004
N Kratochwil et al (2020) IEEE TRPMS 2020.3030483

Classification of events with rise time

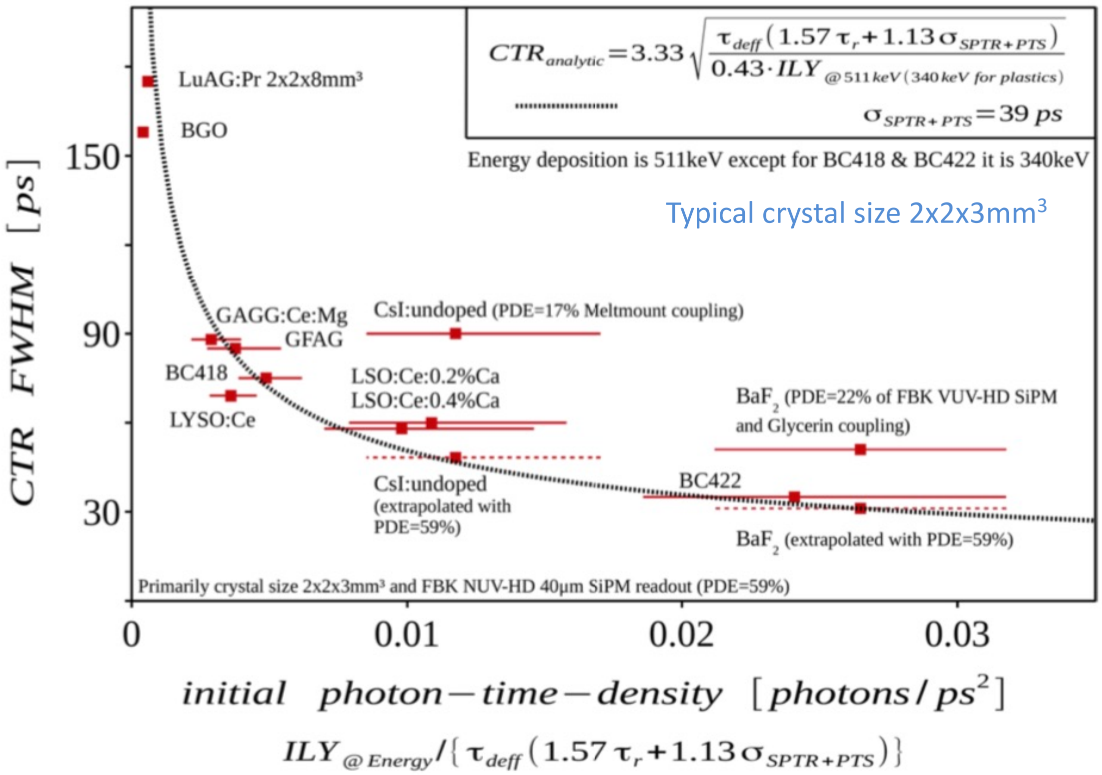






For fastest events CTR of 200ps !

CTR @511 keV for several scintillators



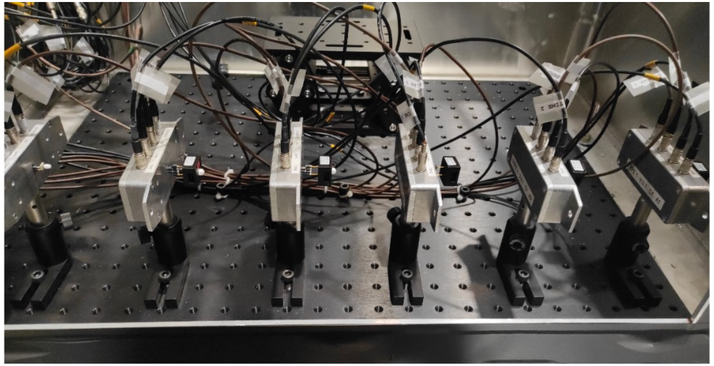
Analytic CTR expression
including SiPM SPTR influence
S. Vinogradov, NIMA 912 (2018) 149-153

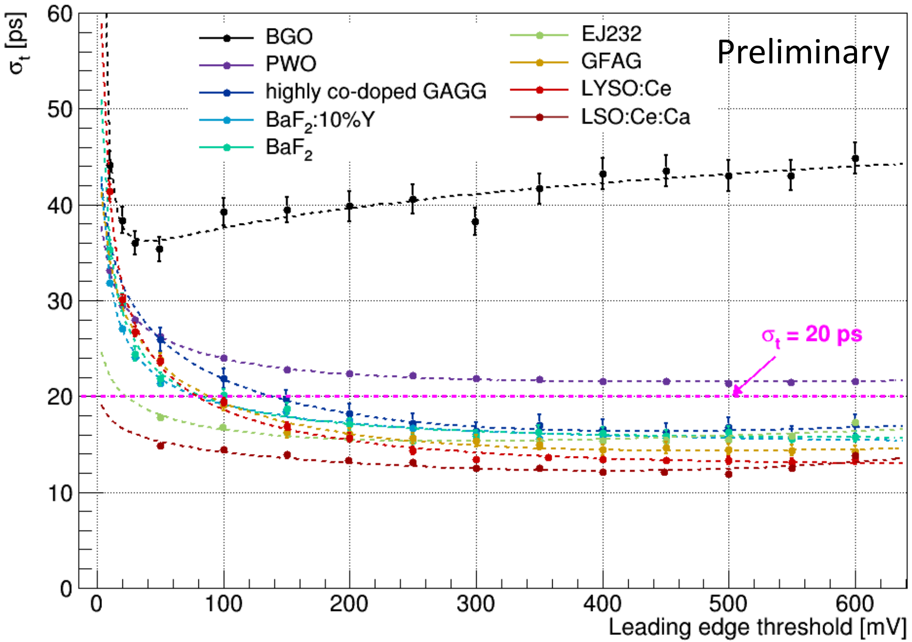
S. Gundacker et al. Phys. Med. Biol. 65 (2020) 025001

Time resolution of several scintillators under mips

Test conditions :

- Scintillator length 10mm except EJ232 (3mm)
- Crystals Teflon wrapped and Meltmount coupled to SiPM
- SiPM used HPK S13360-3050PE SiPMs (except for LSO:Ce:Ca (FBK NUV-HD)
- Readout with HF amplifier



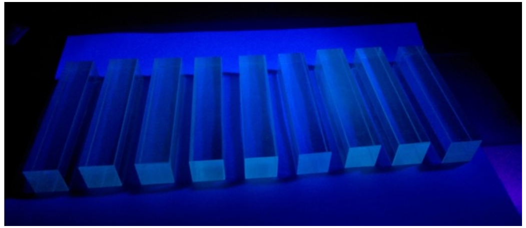


R. Cala' et al., Paper in preparation

Development on Scintillating Glasses

- Scintillating glasses were considered in the 90's for LHC but were not sufficiently radiation tolerant*
  *See for instance E. Auffray *et al, NIM A* **380** (1996) 524-536; P R Hobson *et al* Journal of Non-Crystalline Solids 213-214 (1997) 147-151, S F Shaukat *et al* Journal of Non-Crystalline Solids 244 (1999) 197-204, CMS note)
- Since some years new developments on glasses within different projects (eg ATTRACT project, EIC R&D)
  - Oxyde and Fluoro glasses
    - Attempt to increase the density and the radiation hardness
    - Progress in production scale

Exemple DSB Glasses Intelum

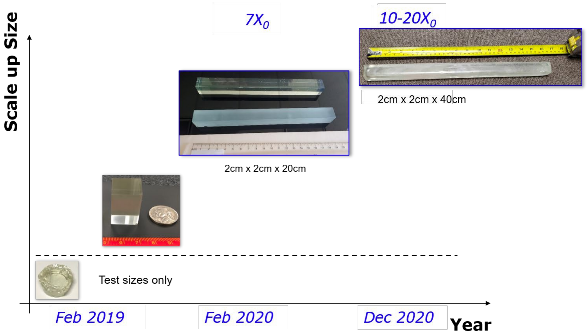


Industrial development via ScintiGlass: Attract project with Preciosa Company

ATTRACT

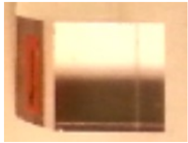
V. Dormenev et al, NIMA, 1015, 2022, 165762

EIC R&D: eRD105 (SciGlass)



From T. Horn, CERN EP R&D, Nov21

Fluorophosphate glasses From AFO company

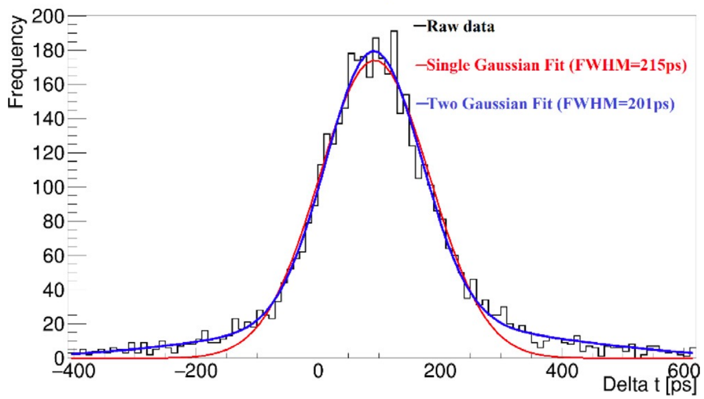


M. Lucchini et al., NIMA A 1051 (2023) 168214

Potential of scintillating glasses for fast timing

DSB Glasses

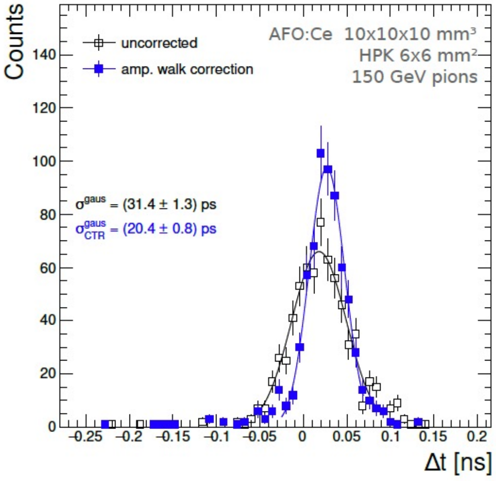
Coincidence time resolution @ 511Kev



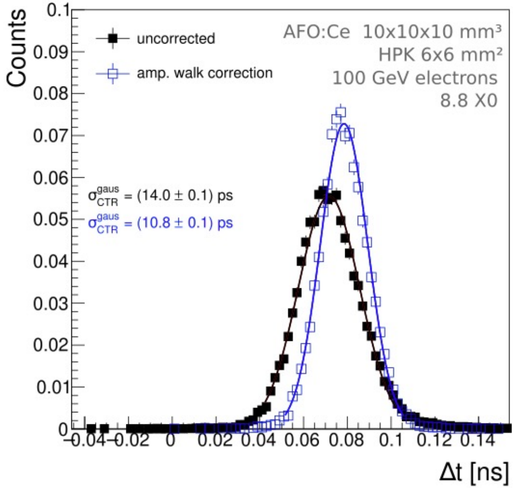
V. Dormenev et al, NIMA, 1015, 2022, 165762

AFO Glasses

Timing resolution with mip



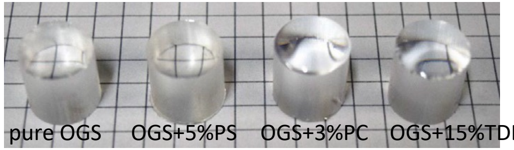
Timing resolution at shower max 100GeV electrons

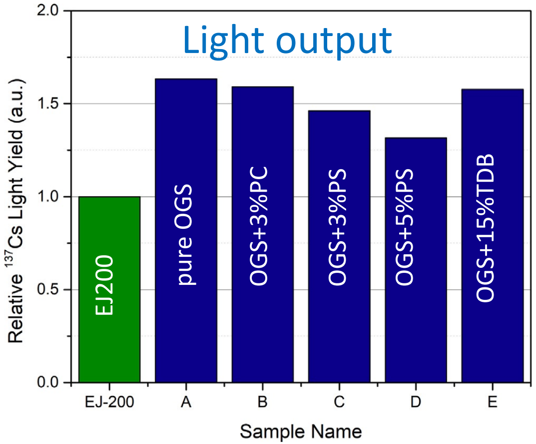


M. Lucchini et al., NIMA A 1051 (2023) 168214

R&D for Organic Scintillators

Organic glasses developed in Sendai National lab



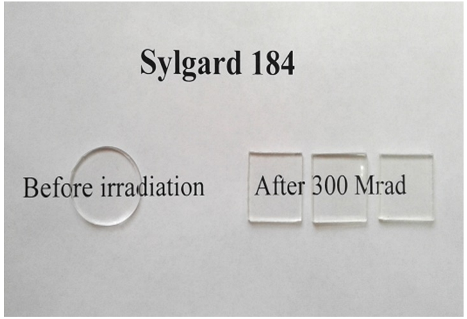


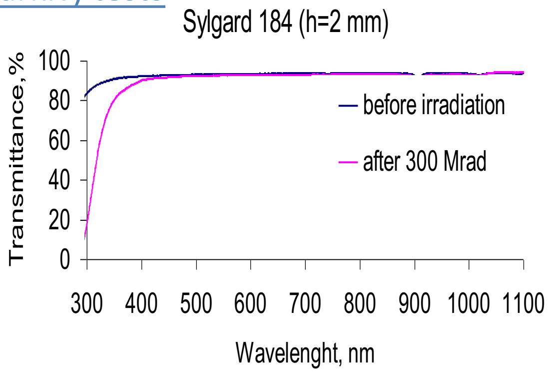
From L. Q Nguyen et al., NIMA 1036 (2022) 166835

Polysiloxane materials

Irradiation with electrons (E_0 = 8.3 MeV) up to 300 MRad dose

ISMA (Kharkiv) tests



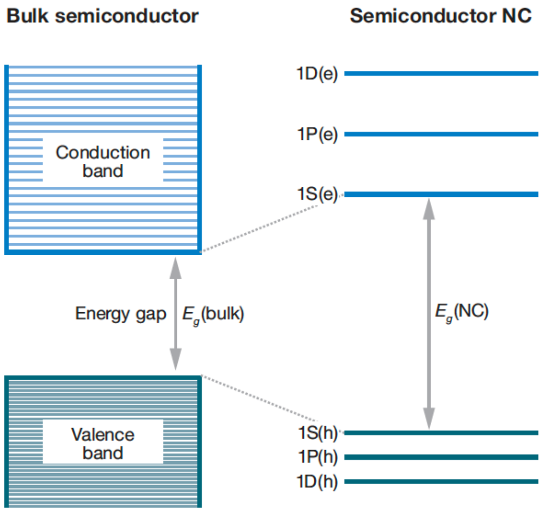


Courtesy A Boyarintsev, ISMA, Kharkiv

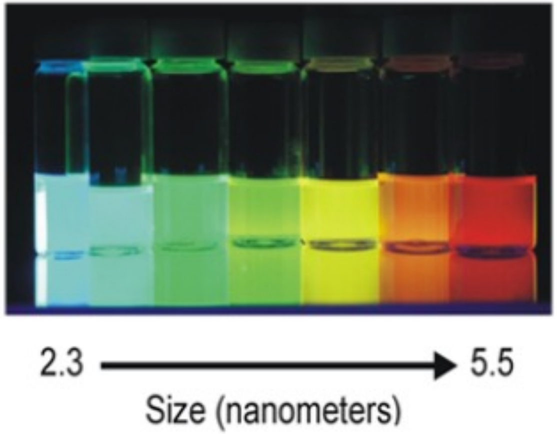
See also A. Boyarintsev NIMA 930, 2019, 180–184
A. Quaranta et al. NIM B, 268, Issue 19, 2010, Pages 3155-3159

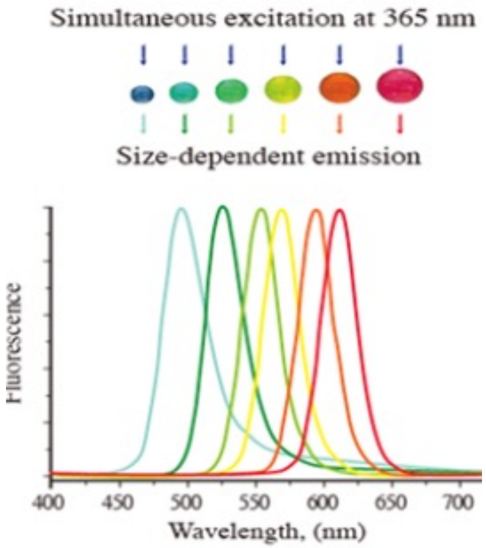
From Bulk to Nanomaterial: Quantum Confinement

Same crystal lattice but nanometer-sized crystal particle



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



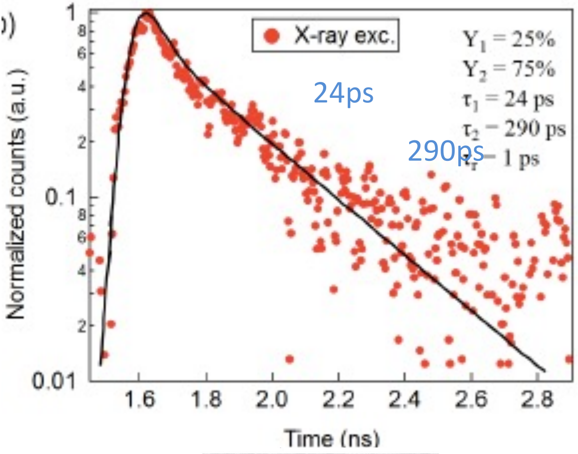


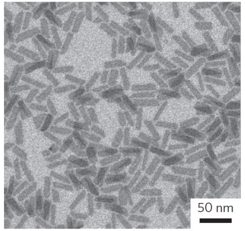
from Benoit Dubertret and Hideki Ooba

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

Subns emission with nanomaterials

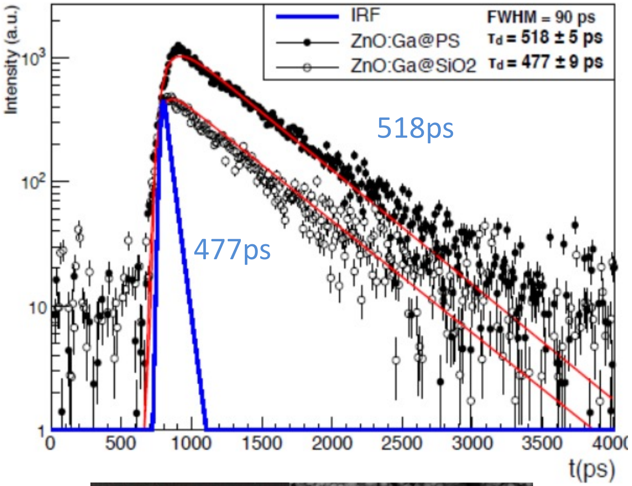
CdSe nanoplatelet,

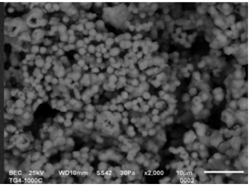




J. Grim et al., *Nature Nanotechnology*, **9**,2014, 891–895
R. Martinez Turtos et al., 2016 JINST_11 (10) P10015

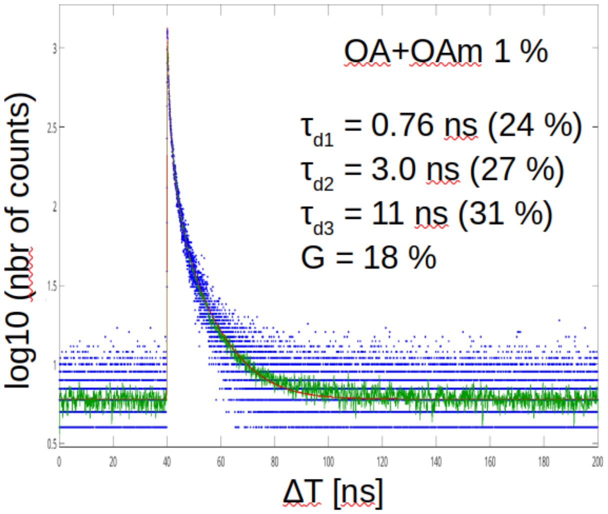
ZnO:Ga embedded in SiO_2 or polystyrene

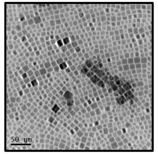




Procházková et al., Radiat Meas 90, 2016, 59-63
R. Turtos Phys. Status Solidi RRL 10, No. 11, 843–847 (2016)

$CsPbBr_3$ embedded in polystyrene

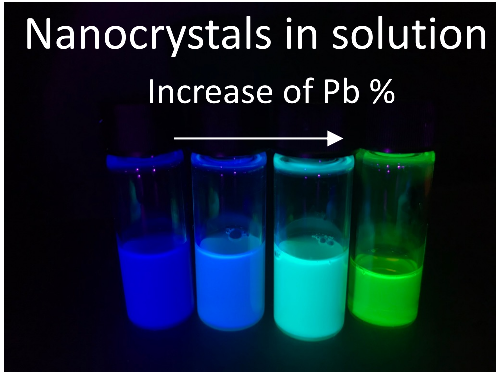




K. Děcká et al. Journal of Material Chemistry C 10(35):12,836–12,843.

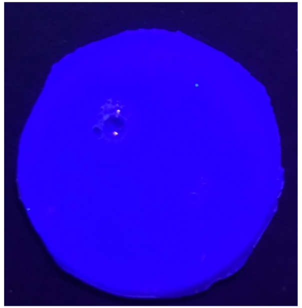
$CsPb(Cl/Br)_3$ Scintillating nanocomposite

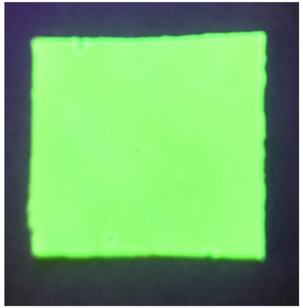
Tunable emission

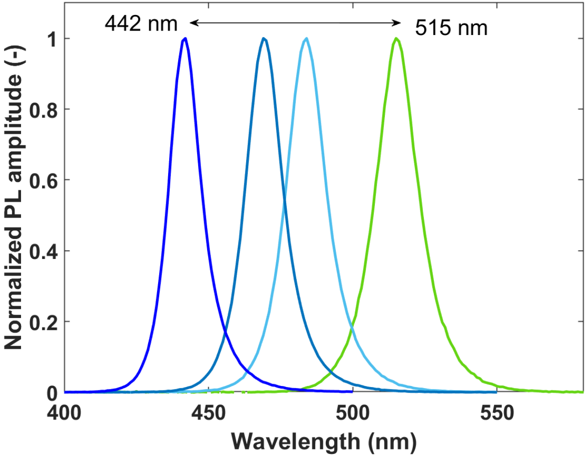


Polymerisation

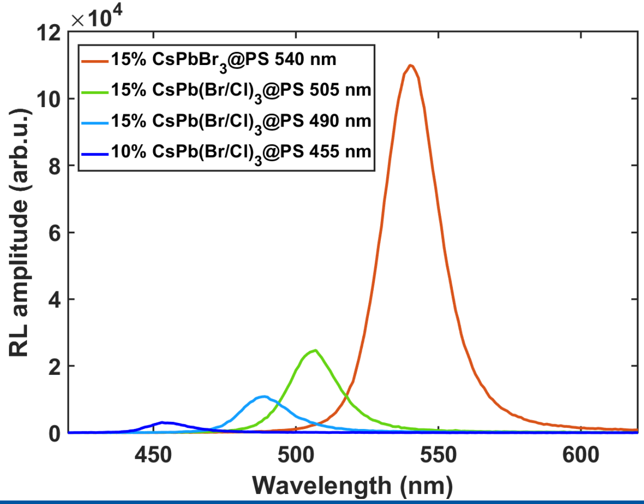
Nanocrystals embedded in polymer





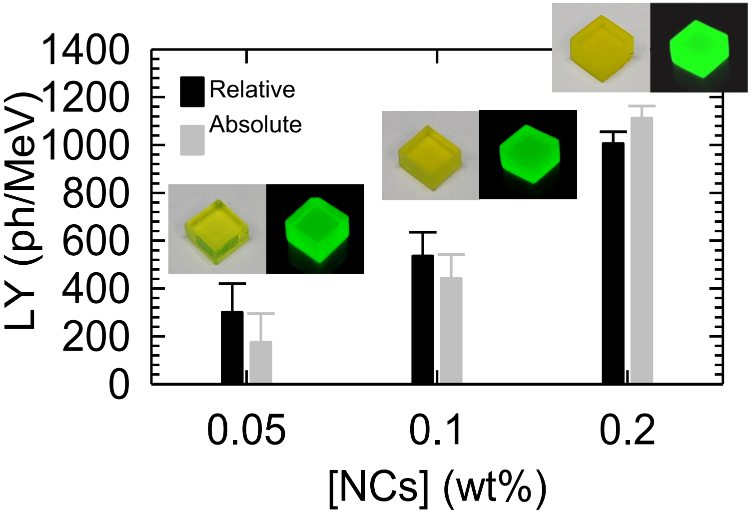


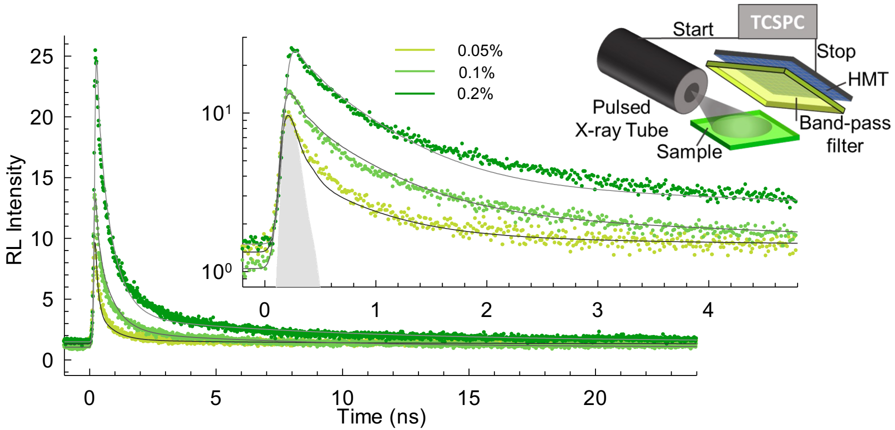
Courtesy J. Kral, CTU, Prague



$CsPbBr_3$ Scintillating nanocomposite

R&D to increase concentration of nanomaterial in the host





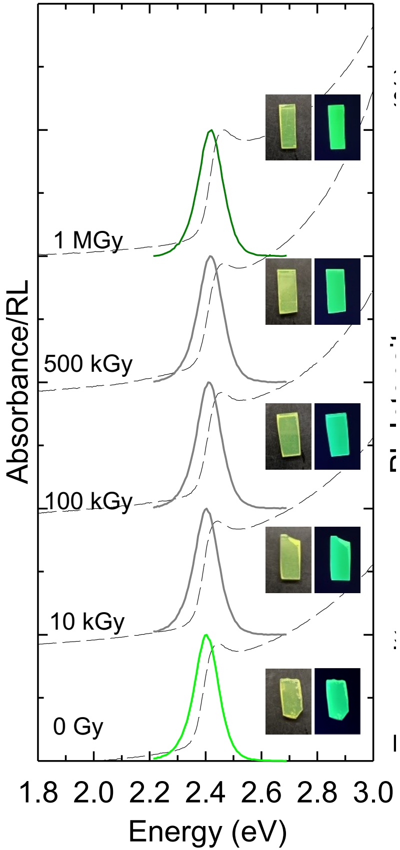
[NC] (wt %)	Pro mpt	t_1		t_2		t_{EFF}	CTR
	R_P	R_1	ns	R_2	ns	ns	Estimated (ps)
0.05	0.30	0.37	0.61	0.33	22	1.13	93
0.1	0.32	0.21	0.62	0.47	8.7	1.76	81
0.2	0.34	0.22	0.60	0.44	6.8	1.54	51

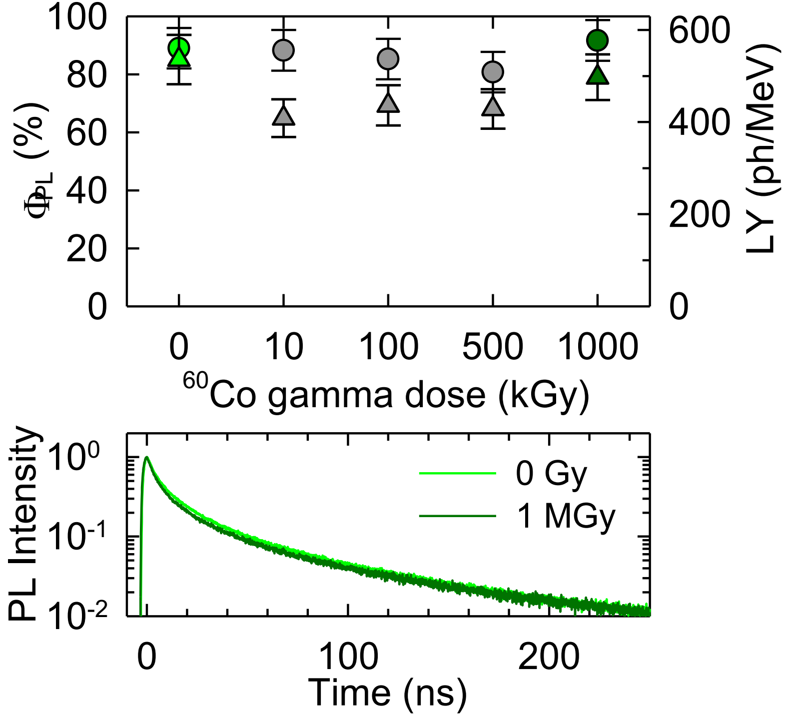
A. Erroi et al., in press in ACS Energy lerters

Very fast emission

$CsPbBr_3$ Scintillating nanocomposite

Radiation damage study



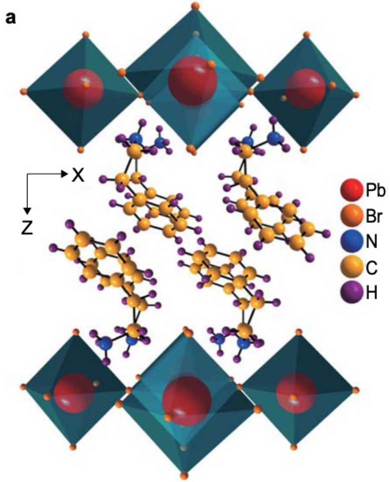


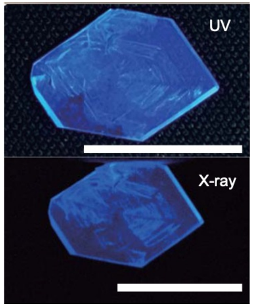
No impact after irradiation with up to MGy

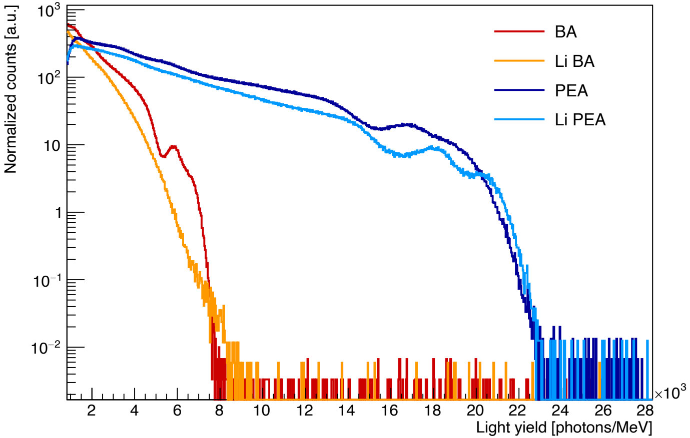
A. Erroi et al., in press in ACS Energy letters

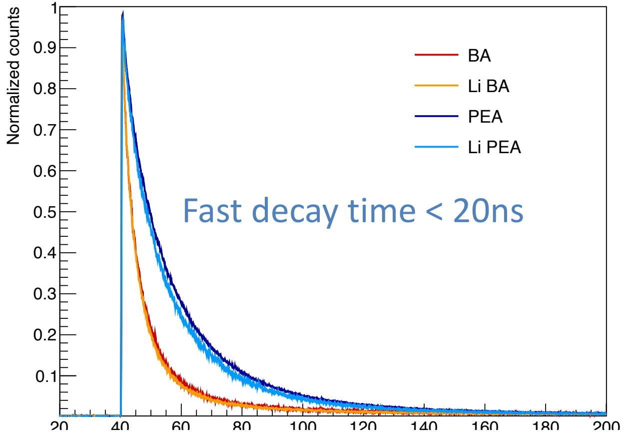
Two dimensional hybrid perovskites

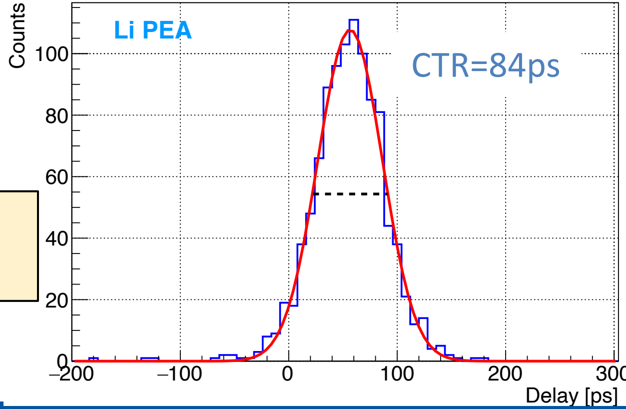
An organic-inorganic hybrid structure.









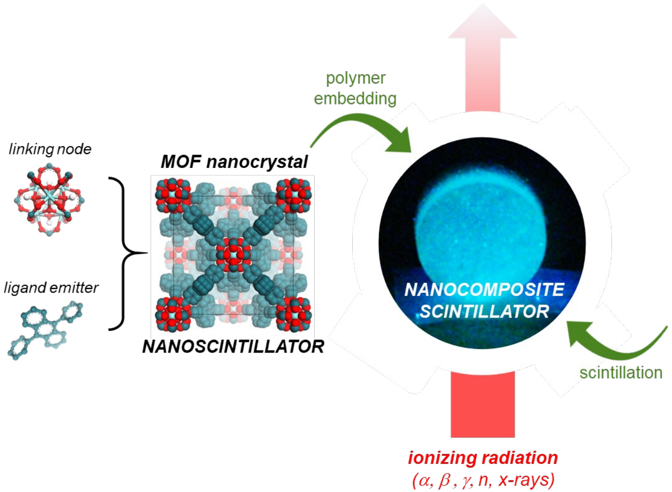


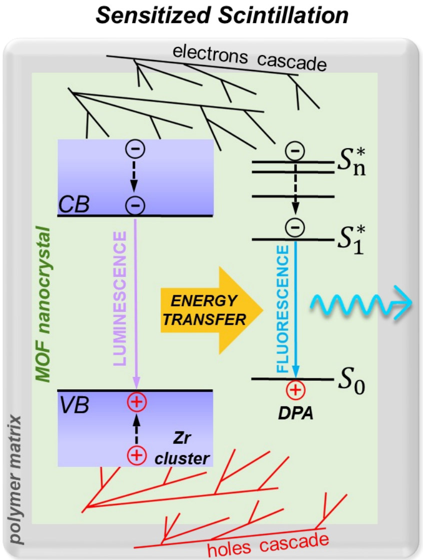
Relative high light output 20000ph/MeV for PEA type and good timing properties

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

R. Cala et al, Appl. Phys. Lett. 120 (2022) 241901

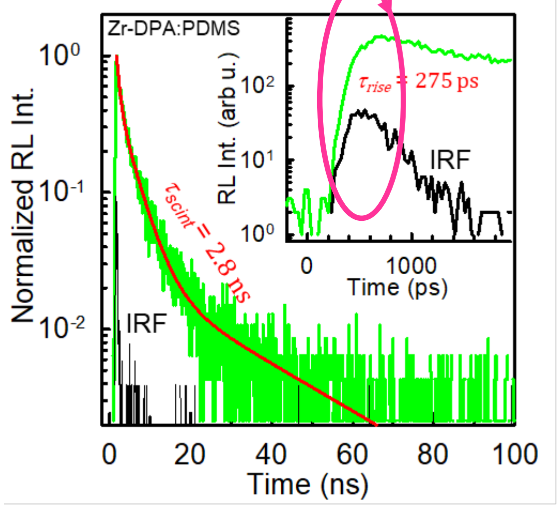
Composite fast scintillators based on high-Z fluorescent metal–organic framework (MOF) nanocrystals





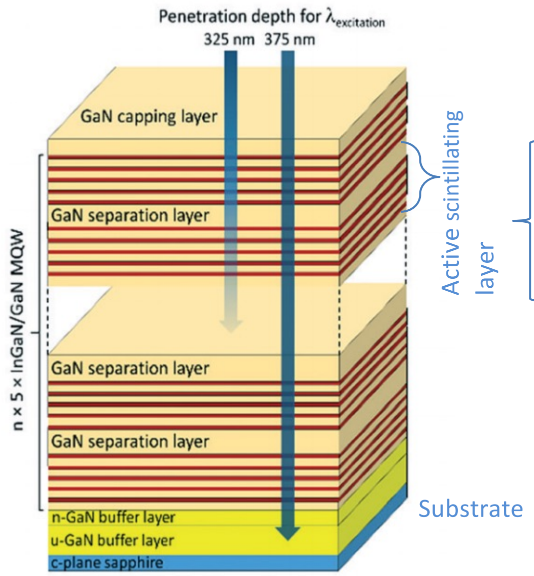
Rise time limited by the instrumental response function

REAL t_{rise} << 300 ps



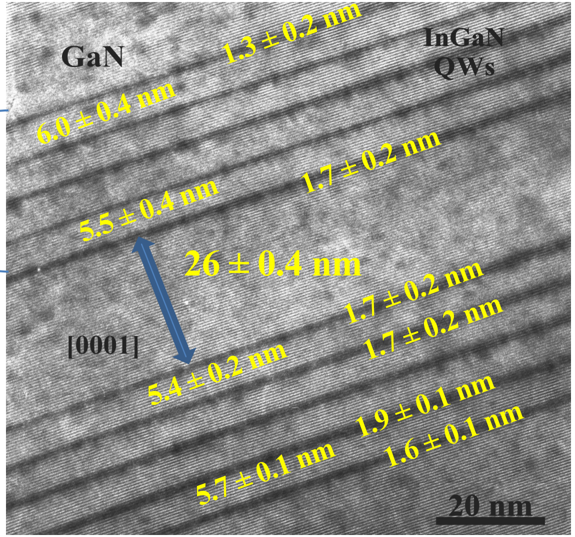
J. Perego, et al. Nat. Photonics (2021). https://doi.org/10.1038/s41566-021-00769-z

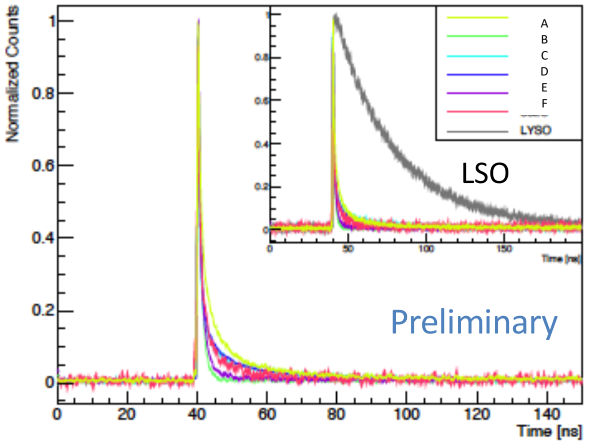
InGaN/GaN heterostructure: Multiple Quantum Wells



T. Hubacek, CrystEngComm, 2019, 21, 356

Picture from A. Hospodkova





F.Pagano, et al, CERN &FZU

Sub-ns fast emisison

FZU Fyzikální ústav Akademie věd České republiky

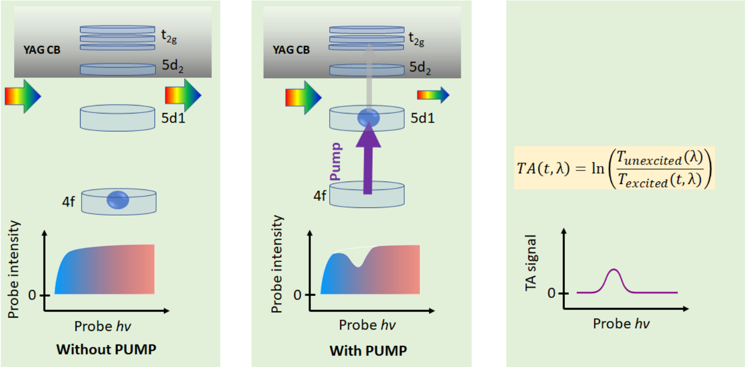
crytur

NEW INSTRUMENTATIONS TO STUDY MATERIALS

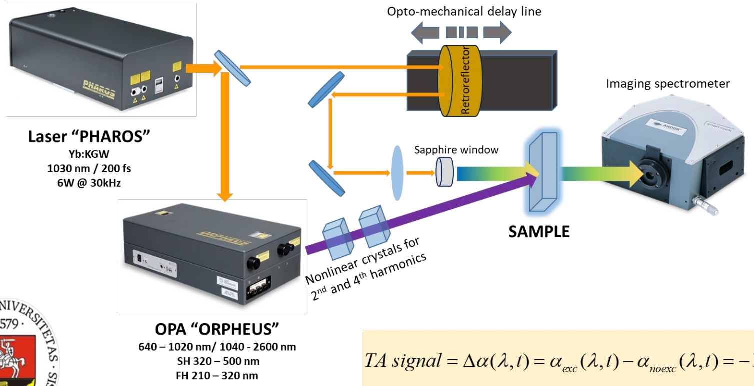
Transient absorption technique

Pump-probe experiment:

- First short laser pulse (pump) temporarily modifies material optical properties
- Second laser pulse (probe) probes this modification by altered transmittivity (absorption)
- By changing the time delay between pump and probe pulses, the modification evolution in time can be traced



$$TA(t,\lambda) = \ln\left(\frac{T_{unexcited}(\lambda)}{T_{excited}(t,\lambda)}\right)$$



$$TA\ signal = \Delta\alpha(\lambda,t) = \alpha_{exc}(\lambda,t) - \alpha_{noexc}(\lambda,t) = -\ln\left[\frac{I_{TR}(exc)}{I_{TR0}(noexc)}\right]$$

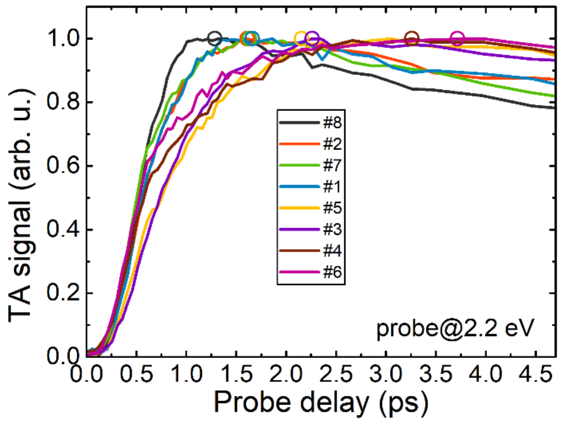
Courtesy G. Tamulaitis, Vilnius university

ADVANTAGES:

- all-optical contactless signal readout
- the time resolution of the measurements is limited just by the laser pulse duration
- enables selective excitation via specific optical transitions targeted in the crystal
- the dependences of pump-induced transient absorption on probe photon energy and time, which are simultaneously obtained in the experiment, facilitate the discrimination of contributions of different kinds of non-equilibrium carriers

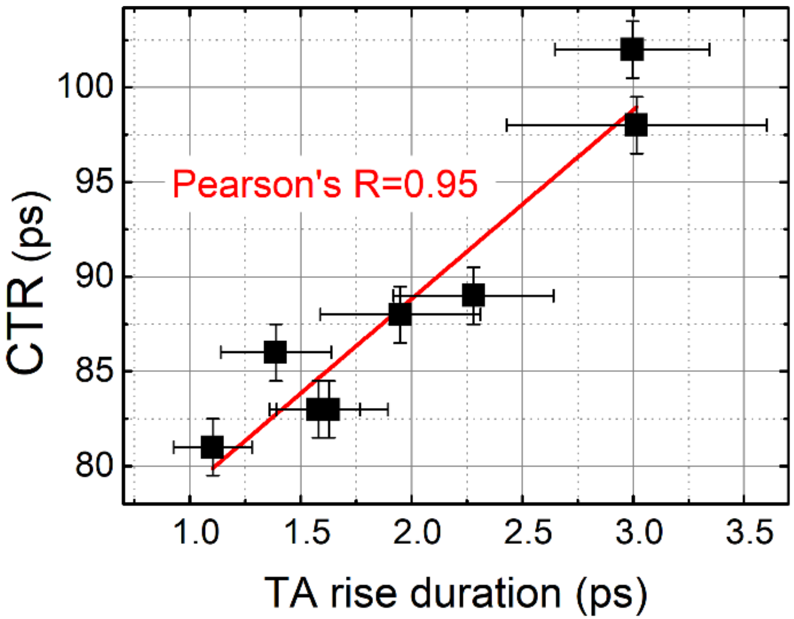
Transient absorption technique: measurement example

TA on LYSO:Ce



Initial part of transient absorption kinetics after excitation with 200 fs pulse in LYSO:Ce samples. The peak of TA response for every kinetics is indicated by a circle of corresponding color.

CTR versus TA rise time



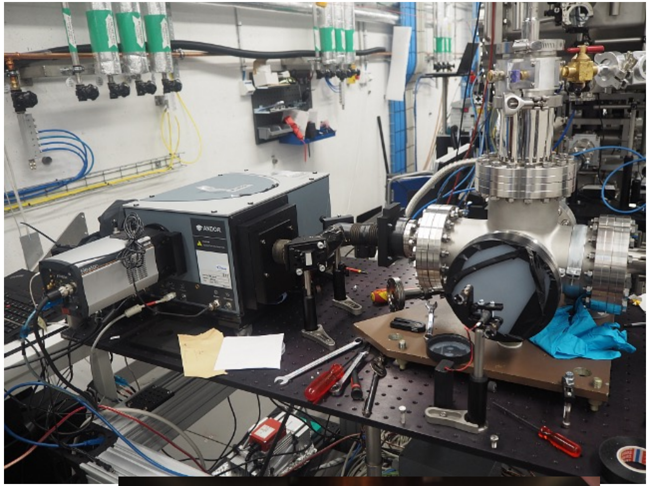
Excellent correlation between the TA rise duration and coincidence time resolution (CTR) is observed
=> TA rise duration is a good figure of merit for the characterization of LYSO:Ce scintillation crystals.

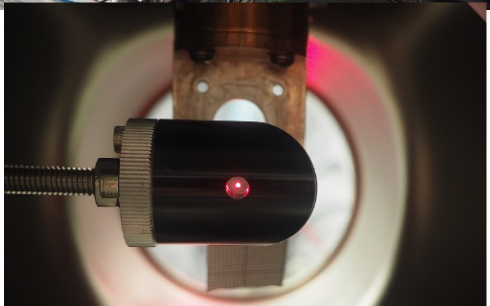
Courtesy G. Tamulaitis, Vilnius university

In more detail:
G. Tamulaitis etal., Radiation Physics and Chemistry 206, 110792 (2023)

Setup for studies of ultrafast (UF) time-resolved luminescence at FemtoMAX in Lund

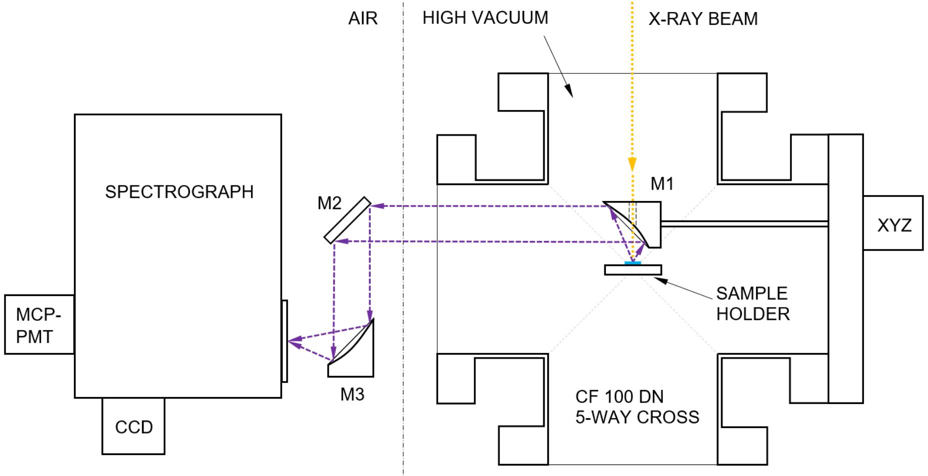
Courtesy M. Kirm, Univ. Tartu





Janis VPF-800 LN cryostat with a sample
oldermounted on the top port of the cross

Linac + undulator
Up to 10 KeV hard X ray photon, 10 Hz repetition
Xray flux ~ 1.5*10^6 ph/pulse
< 200 fs pulse duration



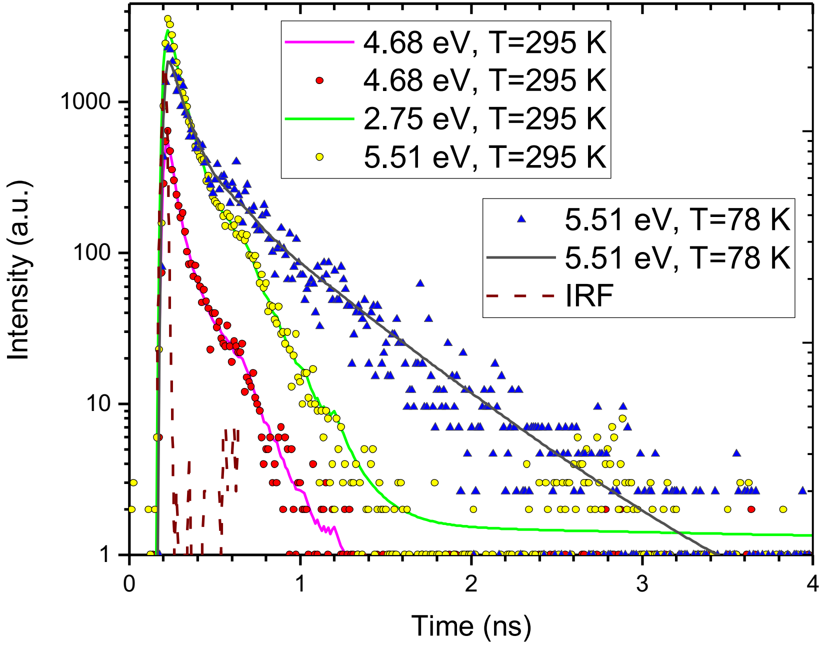
Instrumental response : 32 ps FWHM with MCP-PMT!

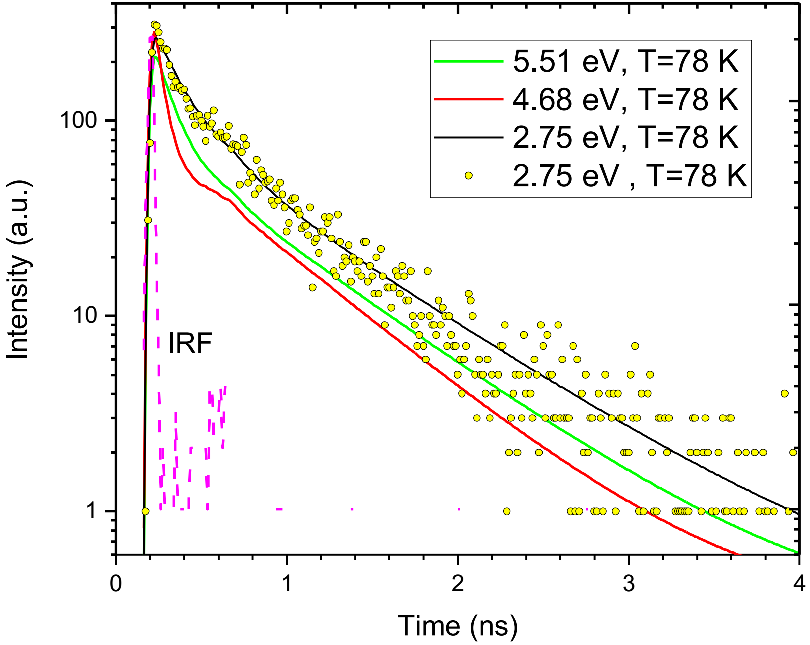
Enquist H, et al. *J Synchrotron Radiat*. 2018;25(Pt 2):570-579.
https://www.maxiv.lu.se/accelerators-beamlines/beamlines/femtomax/

Decay kinetics of spectrally resolved UF emissions in K_2GeF_6 single crystals at FemtoMAX

Courtesy M. Kirm, univ. Tartu

UNIVERSITY OF TARTU



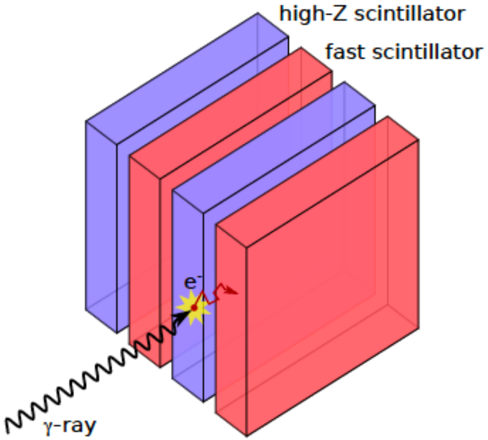


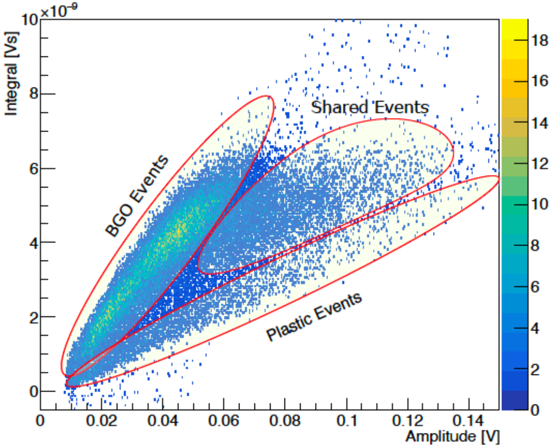
Emission	295 K		78 K	
eV / transition	τ_1 (ps)	τ_2 (ps)	τ_1 (ps)	τ_2 (ps)
5.51 eV K 3p –Ge 4p	71	171	115	699
4.68 eV Ge 4s –F 2p	38	141	50	612

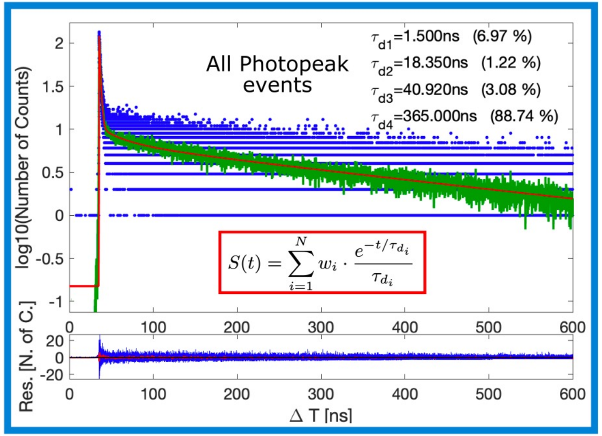
INNOVATIVE CONCEPTS

Heterostructure Concept

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





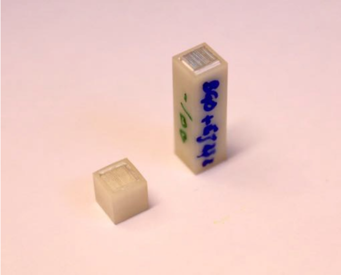


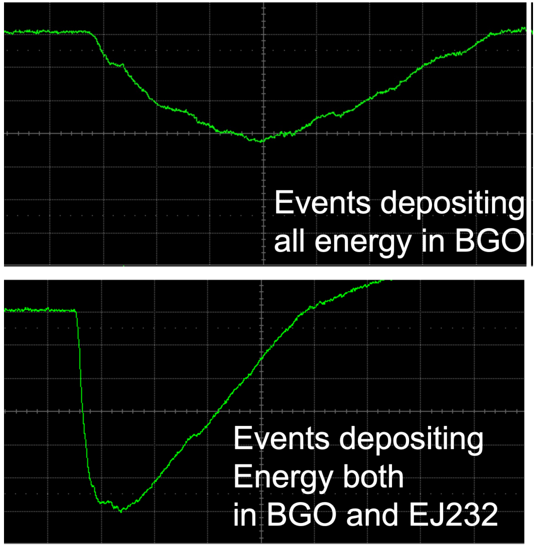
F. Pagano et al, IEEE NSS/MIC2022 under review on TNS

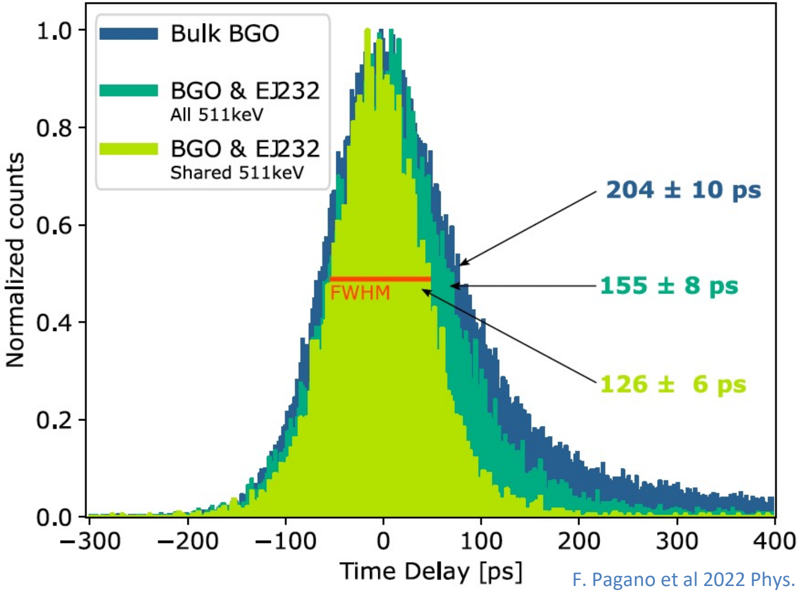
=> Energy sharing between bulk and fast emitter

Concept proposed in the frame of ERC TICAL (GA 338953 PI: P.Lecoq)
R. M. Turtos et al, Phys. Med. Biol. 64 (2019) 85018
F. Pagano et al, 2022, 2022 Phys. Med. Biol. 67 135010

Heterostructure proof of concept with BGO and Plastic



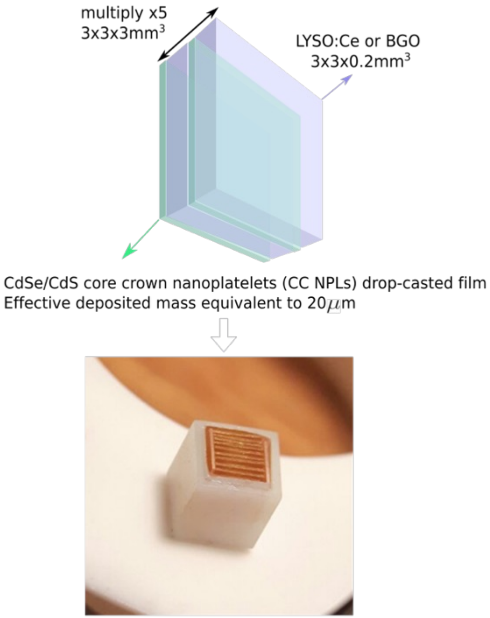


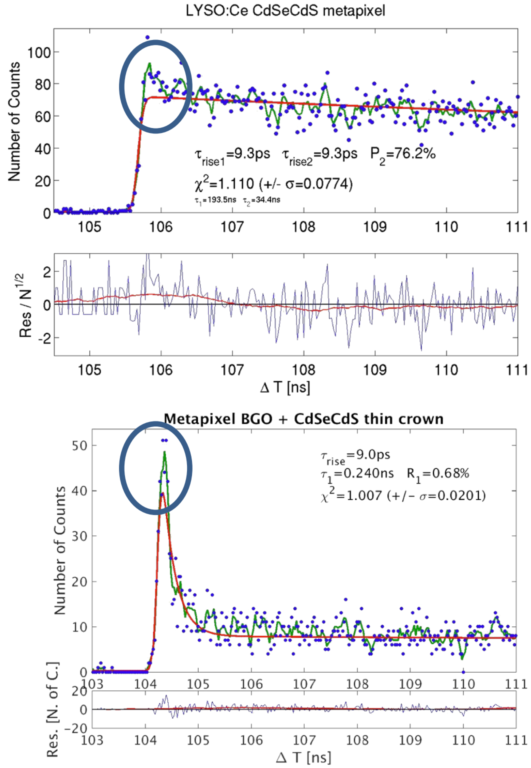


F. Pagano et al 2022 Phys. Med. Biol. 67 135010

Work supported by CERN KT medical applications budget

First attempt of Heterostructure with nanomaterials

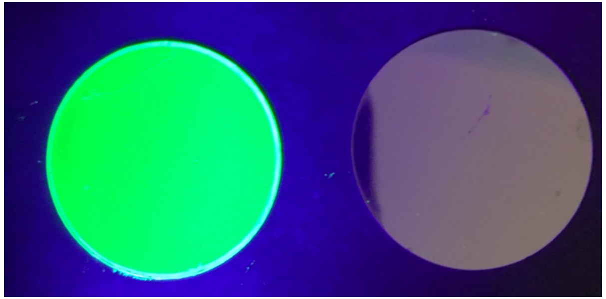




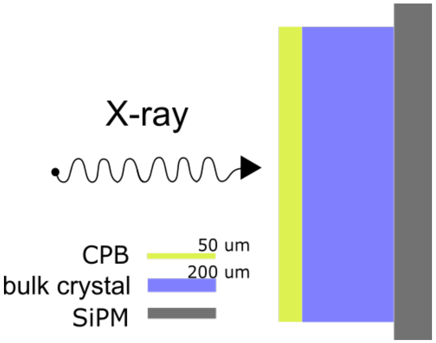
R. M. Turtos et al, Phys. Med. Biol. 64 (2019) 85018
R.M. Turtos, et al. npj 2D Materials and Applications vol. 3, article number: 37 (2019)

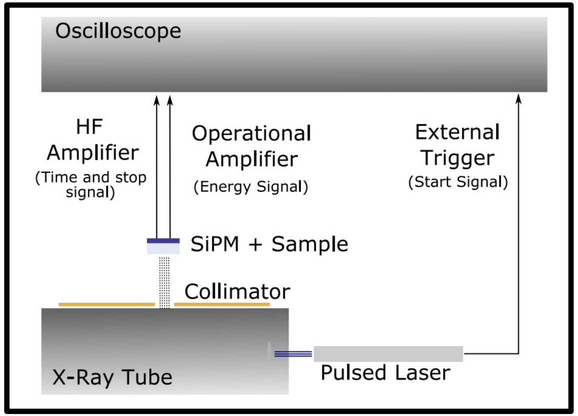
Timing performance of $CsPbBr_3$ nanocrystal layer on bulk GAGG

Thin layer of $CsPbBr_3$ NC on bulk scintillators



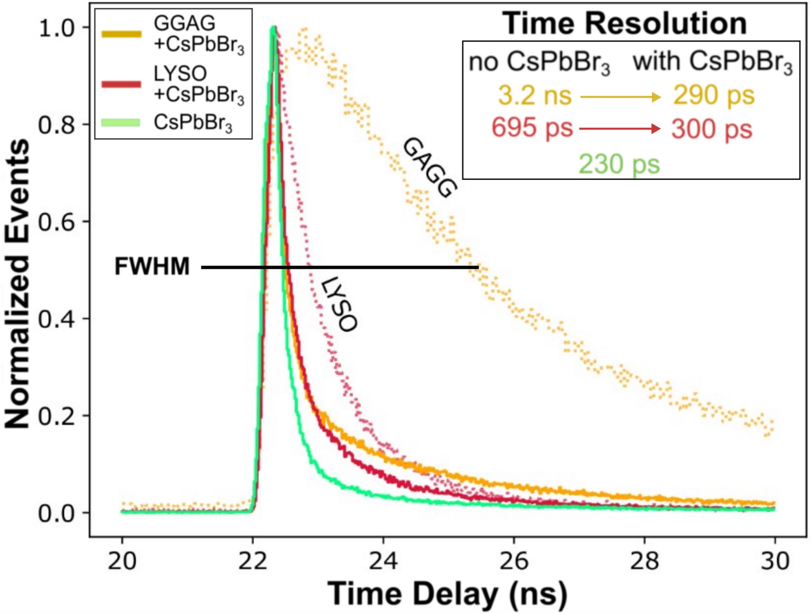
Detector time resolution (DTR) measurements





F. Pagano et al., Frontiers in Physics (vol10)

Detector time resolution with pulsed Xray source



Significant improvements in timing performance under X-ray excitation with nanocrystal layer

Work supported by CERN KT medical applications budget

F. Pagano et al., submitted in Advance Materials Interfaces

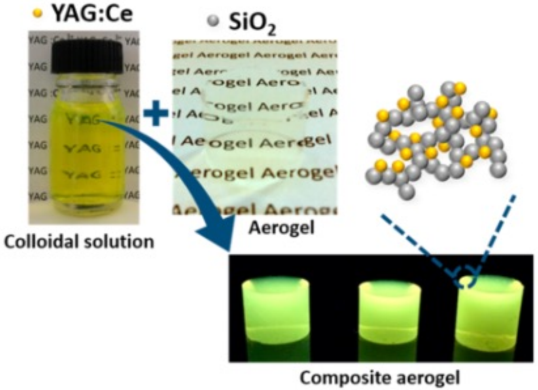
Development of porous scintillators

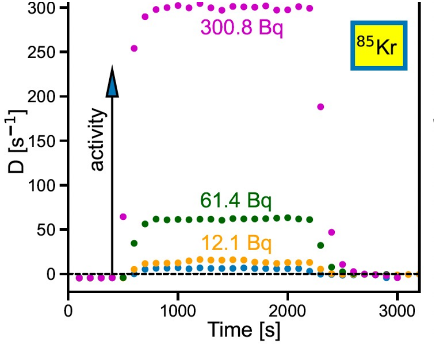
For radioactive gas detection

SPARTE
Scintillating Porous Architectures for RadioacTivE gas detection

Courtesy C. Dujardin
https://www.sparte-project.eu

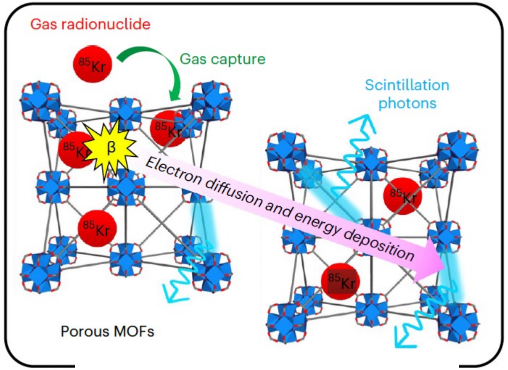
Nanocrystal of YAG:Ce in aerogel

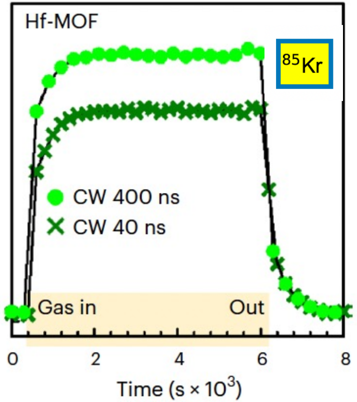




M. Odziomek et al., ACS Appl. Mater. Interfaces 2018, 10, 38, 32304–32312

Scintillating MOF



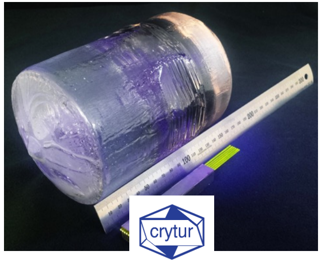


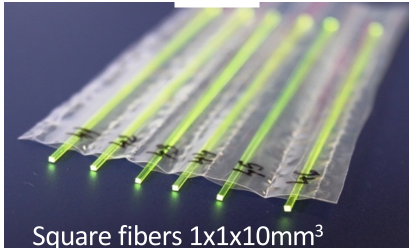
M. Orfano et al., Nature Photonics, 17, 2023, 672–678

New Production Methods

Crystal fibre production

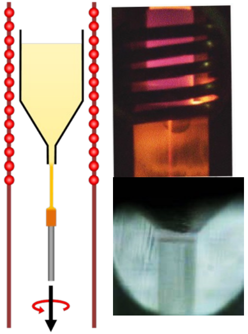
Czochralski method
Fibres cut from large ingot

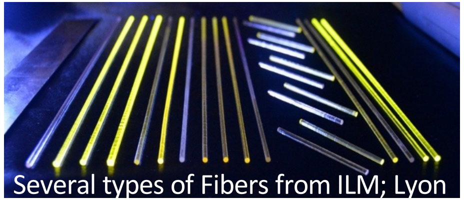




Square fibers 1x1x10mm^3

Micropulling down technique



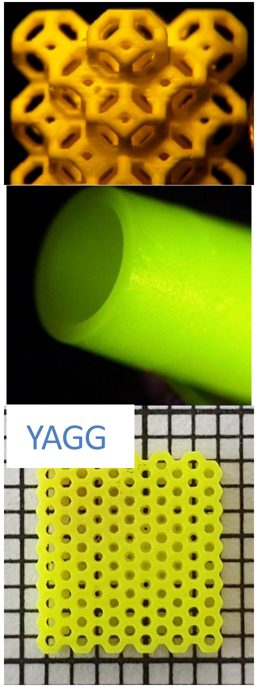


Several types of Fibers from ILM; Lyon

Intelum

3D printing of Scintillators

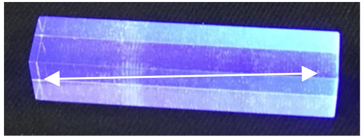
crystals

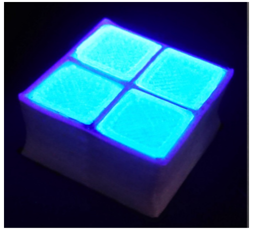


Courtesy of G. Dossovitky, Kurchatov Institute

Plastic scintillator

3D Det project



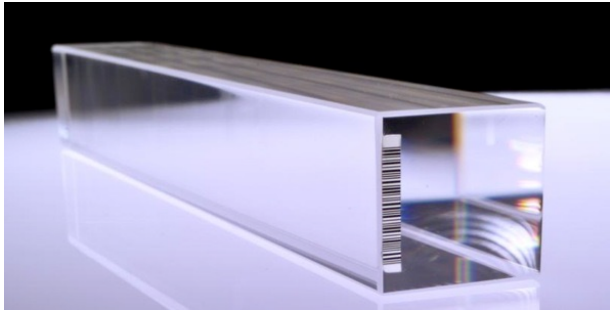


From EP newsletter Nov 21

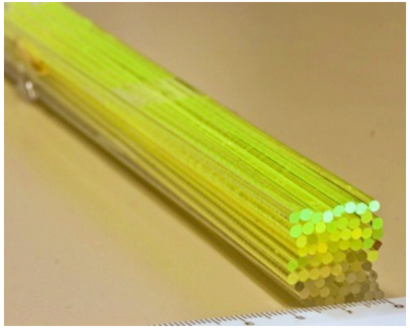
⇒ Feasibility study of crystal fibres production in the ANR project INFHINI and Intelum project (European Rise grant 644260) with 16 Partners (many from CCC) from 12 different countries: 11 academia and 5 companies

Fibres allow flexibility in detector design

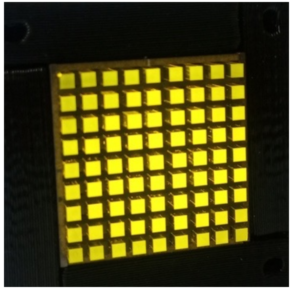
From bulk crystal



To bloc of fibers



To SPACAL



Homogeneous calorimeter

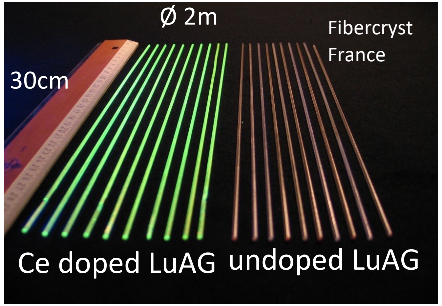
=> Requires large volume of fibres with high density

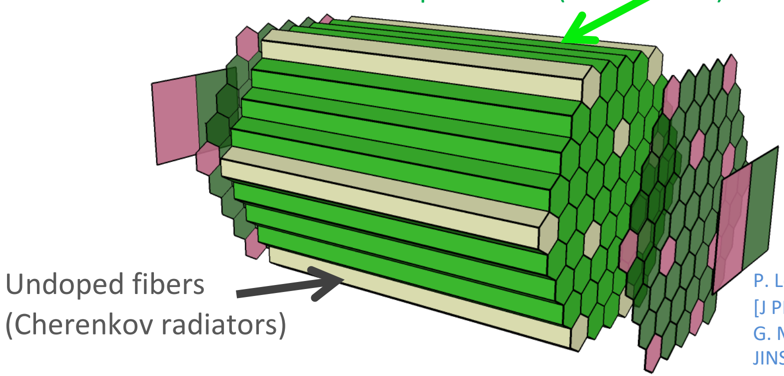
Sampling calorimeter

⇒ requires less fibres, possibility to use materials with lower density

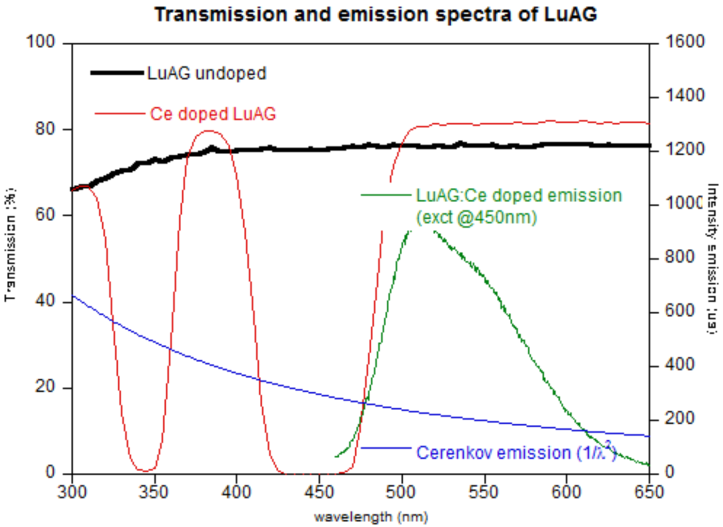
Could be multifunctional: mixed type of fibres
Cerenkov + scintillation +neutrons sensitive
Could play on sampling fraction

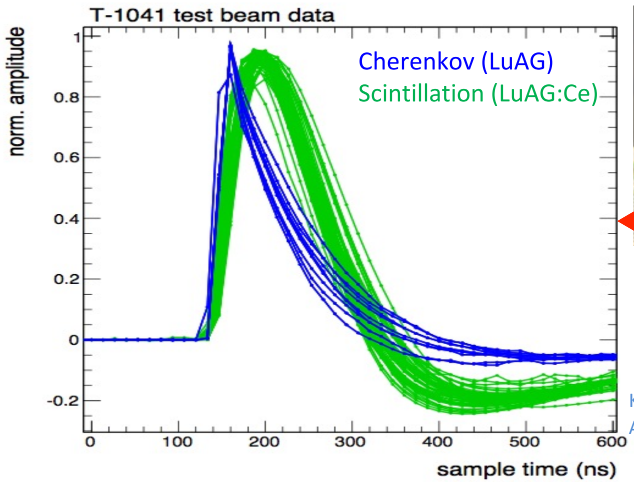
Fibres offer Multifonctionalities

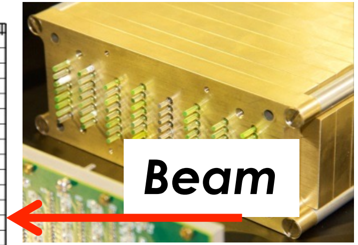




P. Lecoq ,CALOR 2008
[J PHYS 160 (2009) p12016]
G. Mavromanolakis et al. CALOR 2010 +
JINST 6 p10012 (2011)





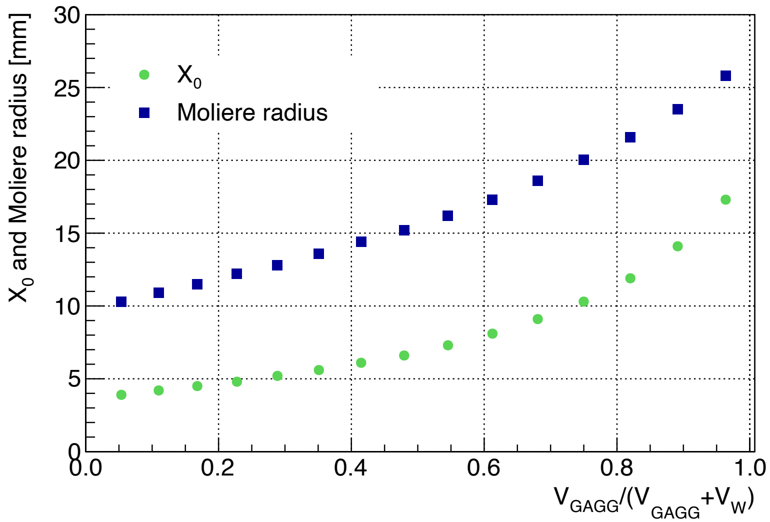


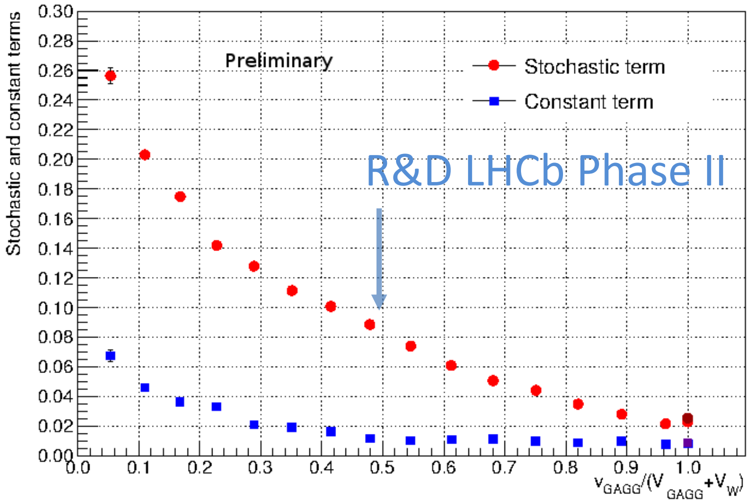
Good separation of Scintillation & Cerenkov

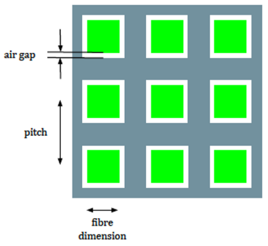
K. Pauwels et al., JINST428 (2013), 8, P09019
A. Benaglia *et al*, JINST 11(5) 05004 (2016)

Tuning of detector performance with SPACAL

Study for :Pitch fixed at 1.67 mm, fibre size variable;







GAGG fibers/W absorber

Modification of Moliere radius and X_0
=> optimisation of granularity

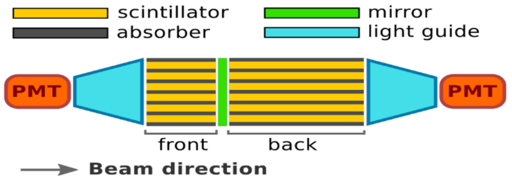
Optimisation of sampling fraction
⇒ optimisation of energy resolution

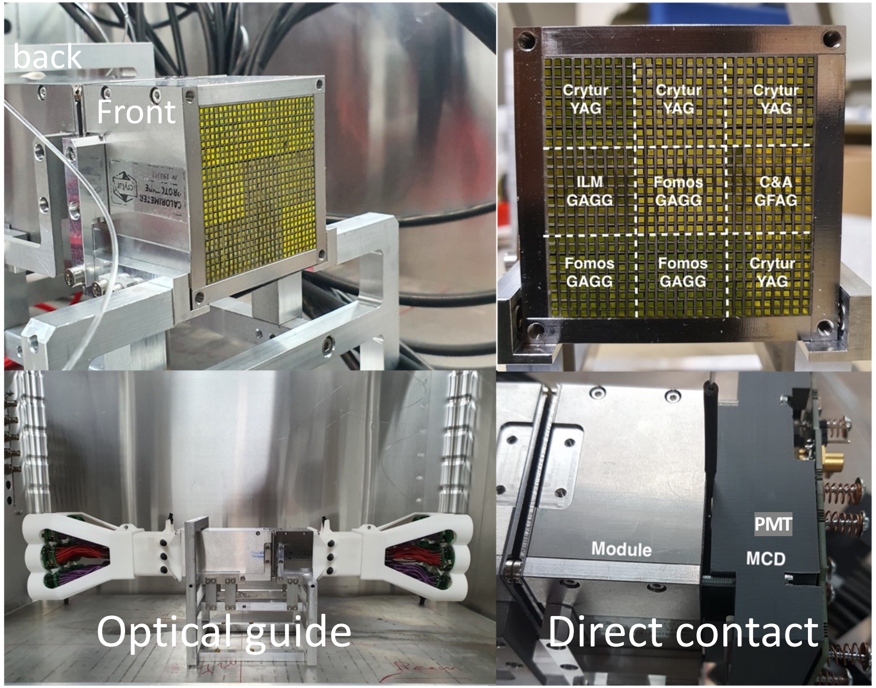
⇒ **R&D on SPACAL with garnet and tungsten in framework of EP_R&D, LHCb upgrade II**

SPACAL-W prototype with garnet crystal fibres

- **Pure tungsten** absorber with 19 g/cm^3 holes with
- **Crystal garnet scintillators**
- 9 cells, each 1.5 x 1.5 cm^2 ($R_M \approx$ 1.45 cm)
- Longitudinal section at the shower maximum
- 4 + 10 cm long split (7+18 X_0), pitch 1.7mm
- Reflective mirror between sections

- Two photodetectors employed:
  - Energy resolution: Hamamatsu R12421 and PMMA light guides
  - Timing resolution: Hamamatsu R7600U-20 metal channel dynodes (MCD) PMTs in direct contact
- 4 garnet types tested:
  - Crytur - YAG
  - Fomos - GAGG
  - ILM - GAGG
  - C&A - GFAG

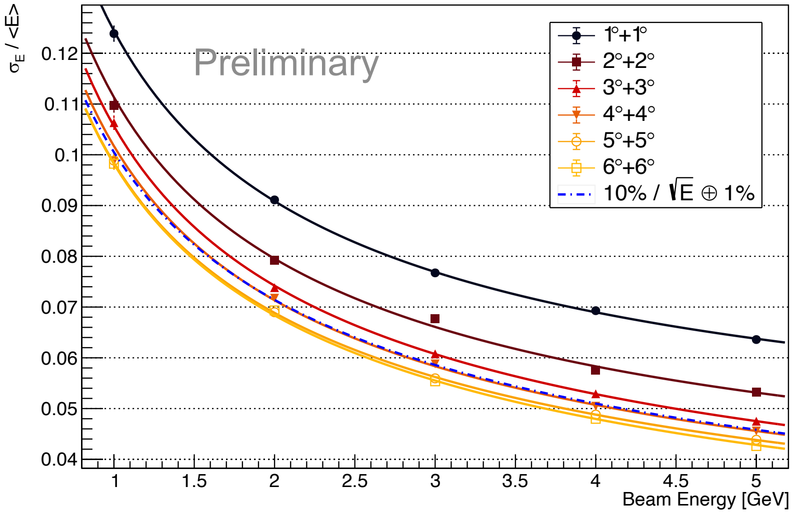




(see talk P. Roloff Monday)

SPACAL-W with garnet crystals: test beam results

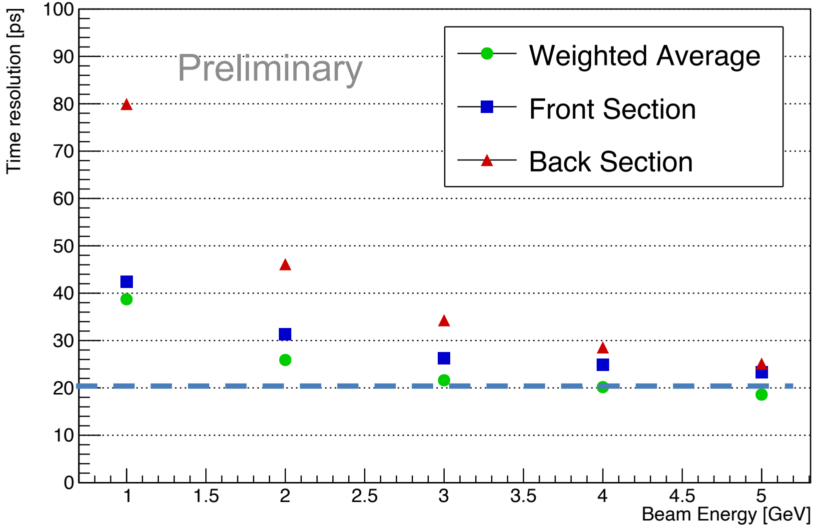
Energy resolution versus incident angles
(DESY 2020 , R12421)



Energy resolution **improves** for larger incident angles

Energy resolution at 3º + 3º
Sampling term: 10.2% ± 0.1
Constant term:1.2% ± 0.3

Time resolution GFAG cell @ incident angle of 3º + 3º
(DESY 2020 , R7600-20)



18 ps @ 5 GeV

(see talk P. Roloff Monday)

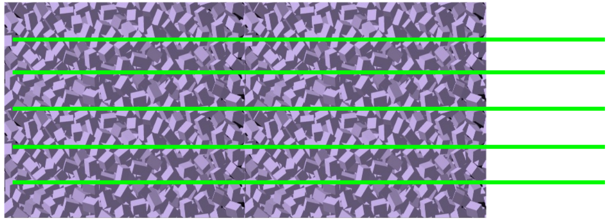
L. Ann, NIM A 1045, 167629 (2022), arXiv:2205.02500

Grainita project

Concept: dispersed submillimetric particles of heavy material ($ZnWO_4$) in dense liquid CH_2I_2 readout with wavelength shifter

$ZnWO_4$ (From ISMA Ukraine):

- LY= 10kph/MeV
- Density 7.62
- Index n=2.1
- τ = 20 μs
- λ_{max} = 480 nm
- grain size : 0.5 mm - 1 mm



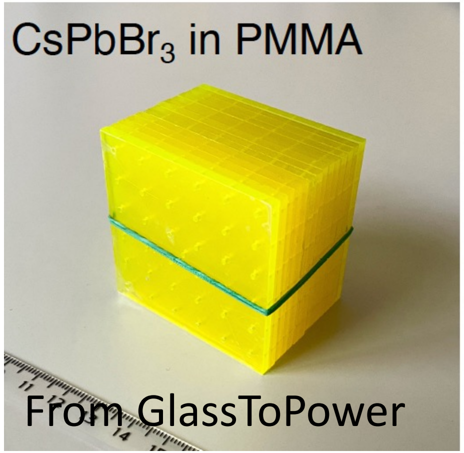
GEANT4 simulation for $ZnWO_4$ + CH_2I_2 cubes (random position) 1mm cubes: $\frac{\sigma_E}{E} \sim \frac{2\%}{\sqrt{E}}$

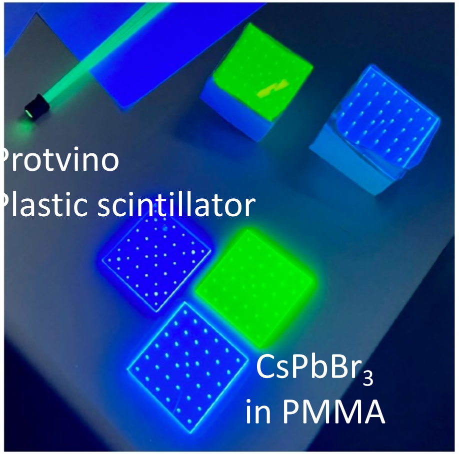
Inspired by LiquidO technique for neutrino detector
(A. Cabrera et al. LiquidO Commun Phys 4, 273 (2021))

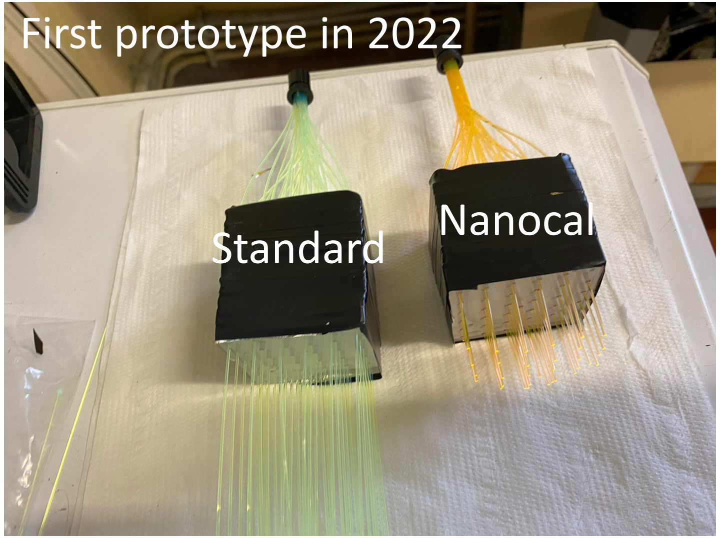
Courtesy M.H. Schune, IJCLab, Orsay, France
on Behalf of Grainita project, see more:
https://indico.in2p3.fr/event/27968/timetable/#20221121.detailed

First Attempt to use Nanomaterial in HEP
Nanocal Bluesky Aidainnova project

Build a Shashlik module with $CsPbBr_3$ nanomaterial embedded in PMMA







Protvino scintillator	NanoCal scintillator
Polystyrene	PMMA
1.5% PTP/0.04% POPOP	0.2% $CsPbBr_3$
Kuraray Y-11(200) fibers	Kuraray O-2(100) fibers

From M. Moulson Aidainnova WP13 20.12.2022

See EP newsletter Nov 22
M. Moulson presentation Aidainnova WP13 20.12.2022

New European Pathfinder Project: UNICORN

Aim to develop nanocomposite scintillator for radiation detector

Consortium of several partners:
UNIMIB, FZU, CERN, ITT, BC materials, Nexdot, Glass to Power,
Starting in June 2023

Conclusion

The field of scintillation is constantly evolving since more than century

Much progress in the understanding of scintillators has been made since the 1990s

The availability of new technologies and methods has enabled a much better understanding of the processes behind

The research on fast emission processes has been strongly fostered by an increasing demand for fast timing detectors

Further R&D is still needed to push the limit:

- Develop bright and fast scintillator:
  - Search for new material
  - Band gap engineering
- Exploit better fast emission process: cross luminescence and Cherenkov emission
  - Will request for better UV sensitive photodetector and optical glue
- Explore the field of quantum confinement

Together with R&D in production methods such as micropulling down, 3D printing, etc..

=> New perspectives for innovative concepts of detectors based on scintillating material with multi-functionalities for next generation of radiation detectors

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ENDO TOFPET US
Endoscopic TOFPET & Ultrasound

TICAL

AIDA 2020

AIDA innova

ATTRACT

Unicorn

THE PICOSEC MC-NET PROJECT

Intelum

ASCIMAT

FAST
Fast Advanced Scintillator Timing

TWISMA

Garnet crystal fibres, Courtesy K. Lebbou, ILM, Lyon, France