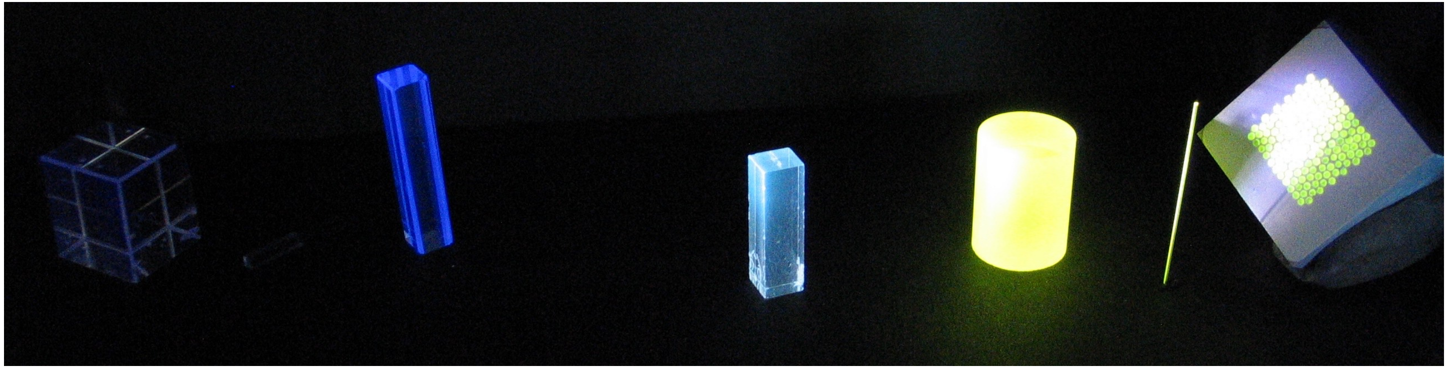
RECENT DEVELOPMENTS IN THE FIELD OF SCINTILLATORS

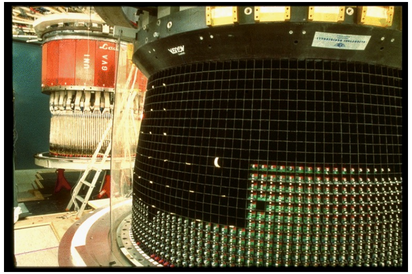
E. Auffray, *CERN, EP-CMX*



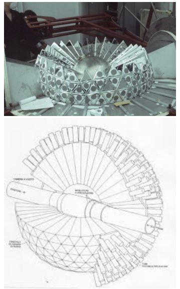
ECFA DRD TF6 calorimetry

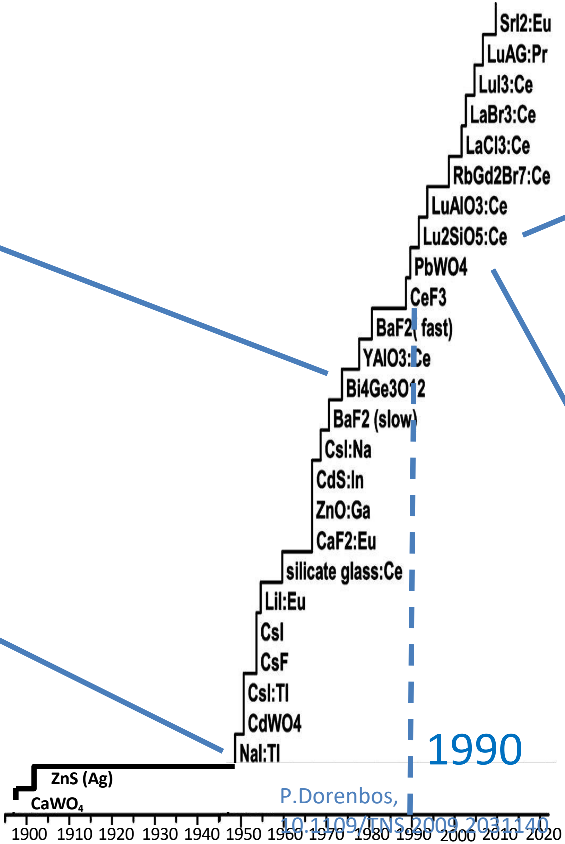
120 years of inorganic scintillators

10752 BGO: L3 calorimeter @LEP 1989



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



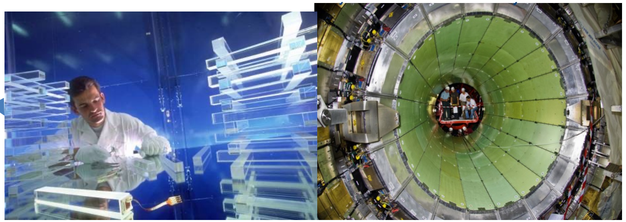


P.Dorenbos, 10.1109/TNS.2009.2031140

1995:LSO: new generation of PET
2026: CMS Barrel timing layer

232000 pixels ~ 3x3X57mm³

75848 PWO: CMS calorimeter @LHC 2008



The Scintillator community

Since more than 30 years a scintillator community exists:

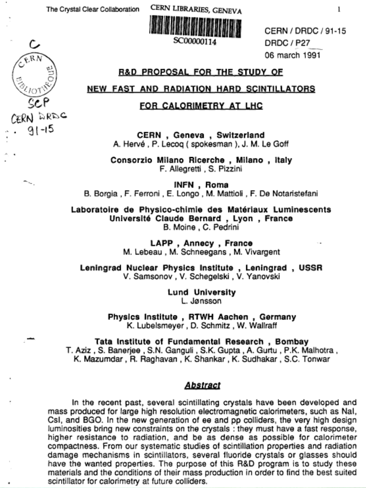
- In 1990 DRDC @ CERN for R&D for future LHC detectors:
  - ⇒ Proposal for develop scintillating materials suitable for use at the future LHC collider, initiated @CERN in 1990 by P. Lecoq, approved in April 1991 by DRDC: RD18
  - ⇒ **Crystal Clear Collaboration**
- In 1992: First conference on inorganic scintillators and their application in Chamonix Crystal 2000
  - => **SCINT community**

Today

Crystal Clear (CERN K1500-TT-PH-004C_CCC): 31 institutes over the world

SCINT conferences every 2 years (http://scint.univ-lyon1.fr/):

Last one in Santa Fe in Sept2022, next one in Milano: June 30-July 5 2024



The Crystal Clear Collaboration   CERN LIBRARIES, GENEVA   1

SC00000114

CERN / DRDC / 91-15
DRDC / P27
06 march 1991

R&D PROPOSAL FOR THE STUDY OF NEW FAST AND RADIATION HARD SCINTILLATORS FOR CALORIMETRY AT LHC

CERN , Geneva , Switzerland
A. Hervé , P. Lecoq (spokesman), J. M. Le Goff

Consorzio Milano Ricerche , Milano , Italy
F. Allegretti , S. Pizzini

INFN , Roma
B. Borgia , F. Ferroni , E. Longo , M. Mattioli , F. De Notaristefani

Laboratoire de Physico-chimie des Matériaux Luminescents Université Claude Bernard , Lyon , France
B. Moine , C. Pedrini

LAPP , Annecy , France
M. Lebeau , M. Schneegans , M. Vivargent

Leningrad Nuclear Physics Institute , Leningrad , USSR
V. Samsonov , V. Schegelski , V. Yanovski

Lund University
L. Jønsson

Physics Institute , RTWH Aachen , Germany
K. Lubelsmeyer , D. Schmitz , W. Wallraff

Tata Institute of Fundamental Research , Bombay
T. Aziz , S. Banerjee , S.N. Ganguli , S.K. Gupta , A. Gurtu , P.K. Malhotra , K. Mazumdar , R. Raghavan , K. Shankar , K. Sudhakar , S.C. Tonwar

Abstract

In the recent past, several scintillating crystals have been developed and mass produced for large high resolution electromagnetic calorimeters, such as NaI, CsI, and BGO. In the new generation of ee and pp colliders, the very high design luminosities bring new constraints on the crystals : they must have a fast response, higher resistance to radiation, and be as dense as possible for calorimeter compactness. From our systematic studies of scintillation properties and radiation damage mechanisms in scintillators, several fluoride crystals or glasses should have the wanted properties. The purpose of this R&D program is to study these materials and the conditions of their mass production in order to find the best suited scintillator for calorimetry at future colliders.

Current R&D on scintillators

R&D on all type of scintillators for many applications:

- organic and inorganic

Improve timing performance:

- Better understanding of the scintillation processes
- Engineering of existing materials (mixed materials, co-doping)
- Looking for faster light emission

Improve radiation hardness:

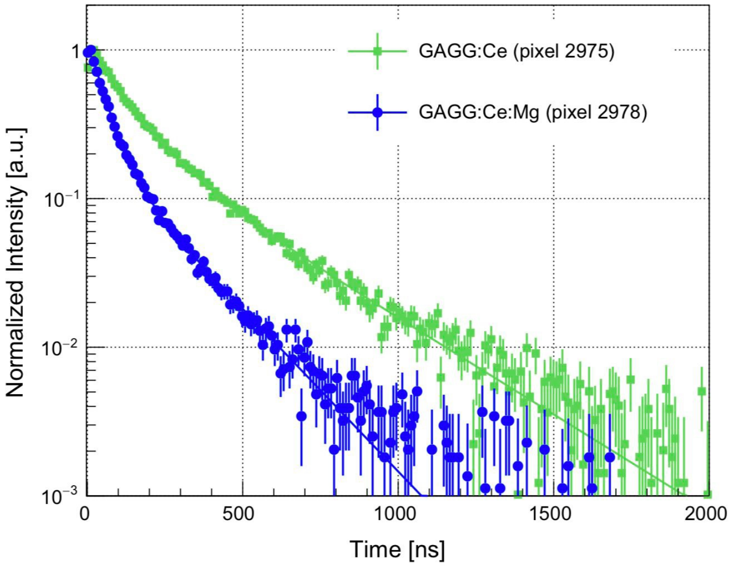
- Understanding of radiation damage mechanisms through tests using different radiation sources (gammas, hadrons, neutrons)
- Scintillator engineering (mixed materials, co-doping):

New production methods

- Crystal fibres
- 3D printing
- Nanomaterial

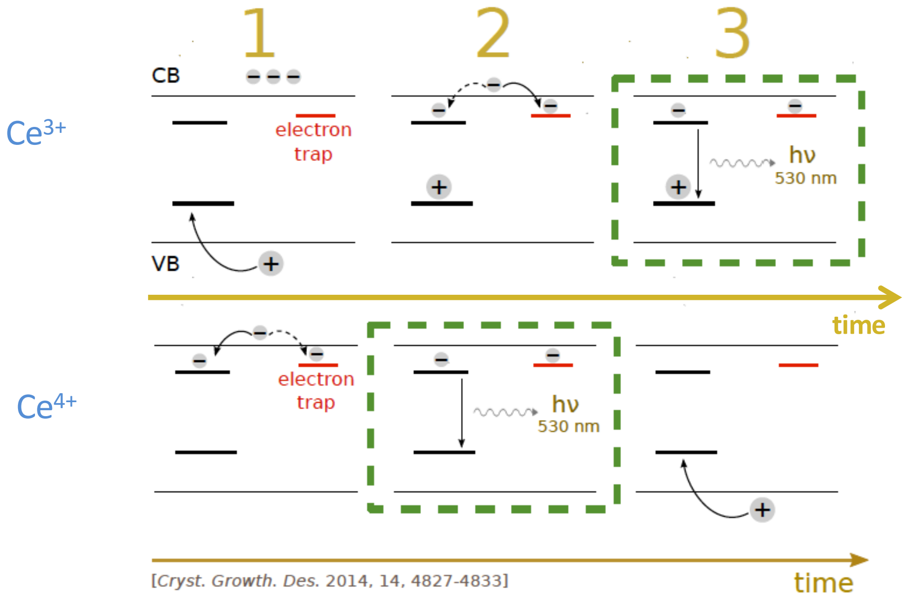
Combination of various materials

Engineering scintillator example of co-doping Ce, Mg in garnet



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

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



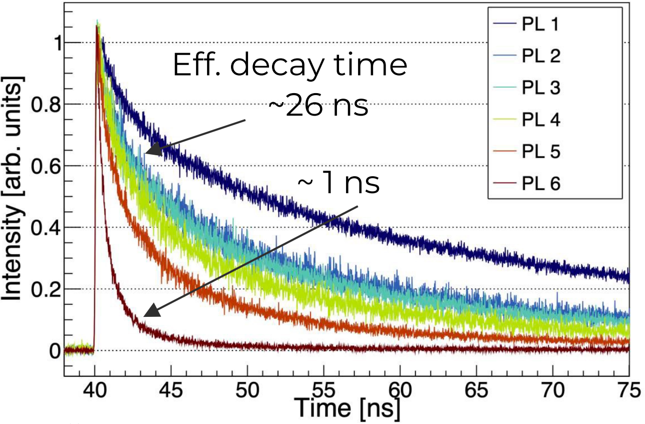
Mg2+ increase Ce^{4+} centers which can directly compete with any electron trap for electron capture in the first instants of scintillator mechanism => Expected faster decay time and lower slow component

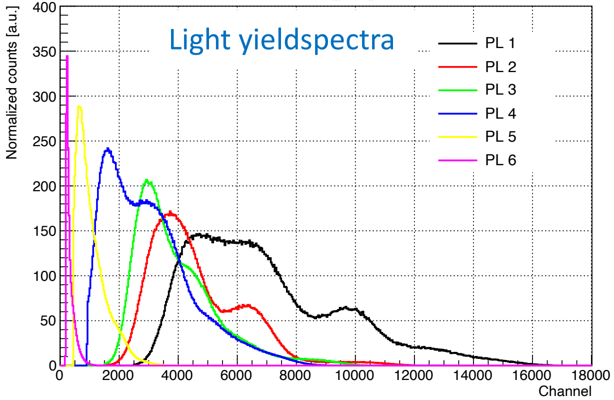
M. Nikl, A. Yoshikawa, Adv. Optical Mater. 2015, 3, 463–481
M. Nikl et al. Cryst. Growth Des. 2014 , 14 , 4827 .

Further acceleration of the emission

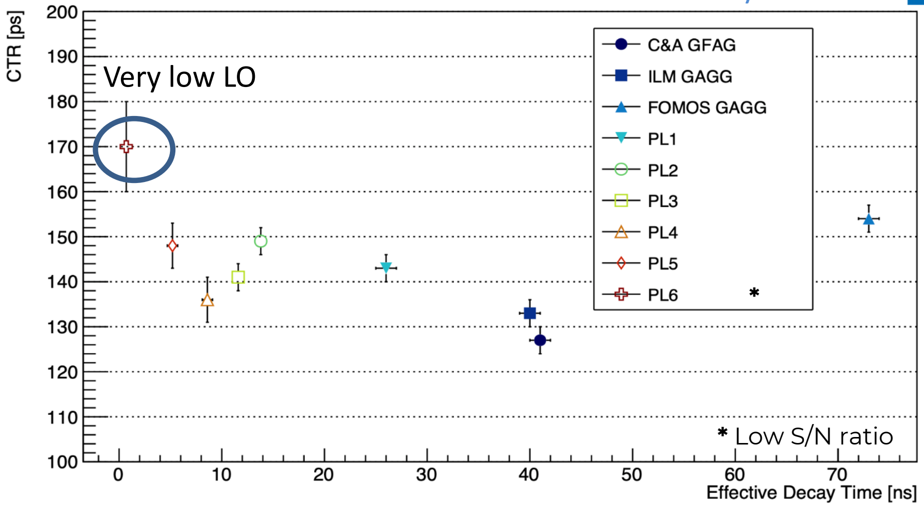
Heavy co-doping Ce^{3+}/Mg^{2+}

Scintillation decay - Pulsed X-Rays





Coincidence time resolution vs effective decay time



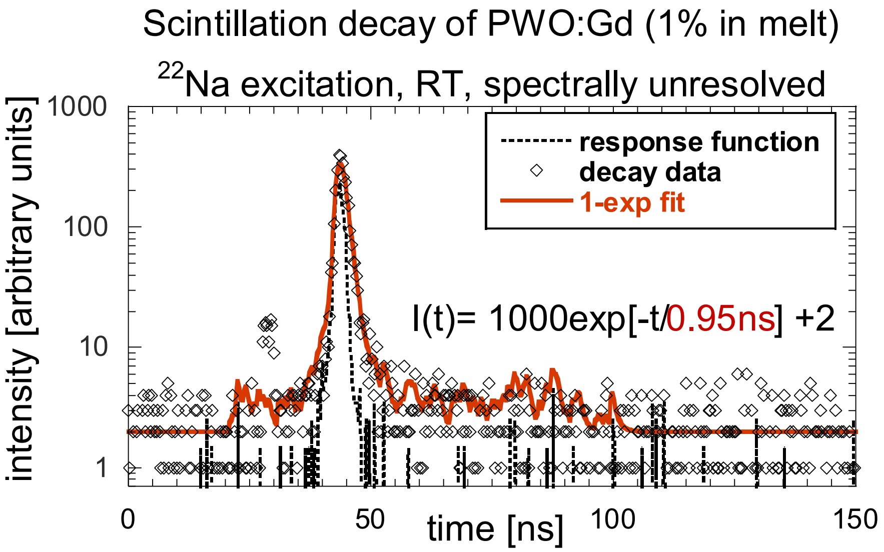
No major loss of time resolution!
Decay time decrease compensated the Light output reduction
=> the same photon time-density

Candidate for LHCb phase II, see talk Philipp today

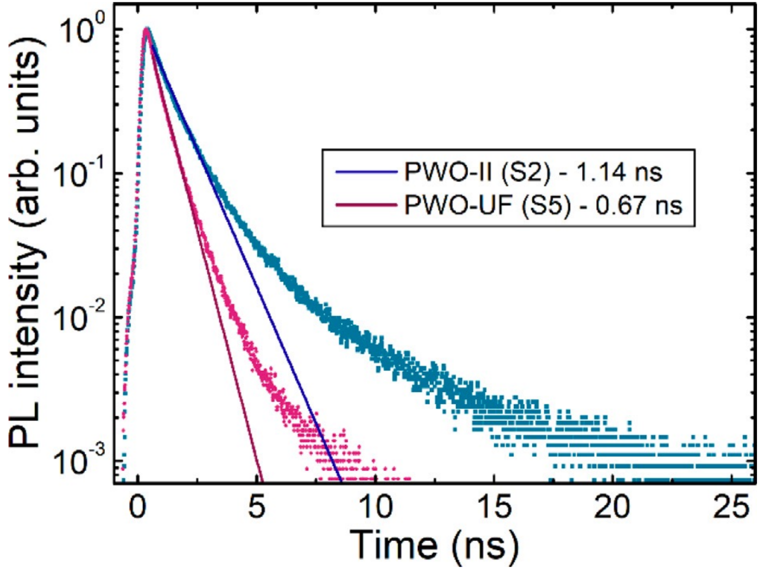
L. Martinazzoli et al., Mater. Adv., 2022, 3, 6842

Towards very fast PWO

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



Photoluminescence decay time @425nm

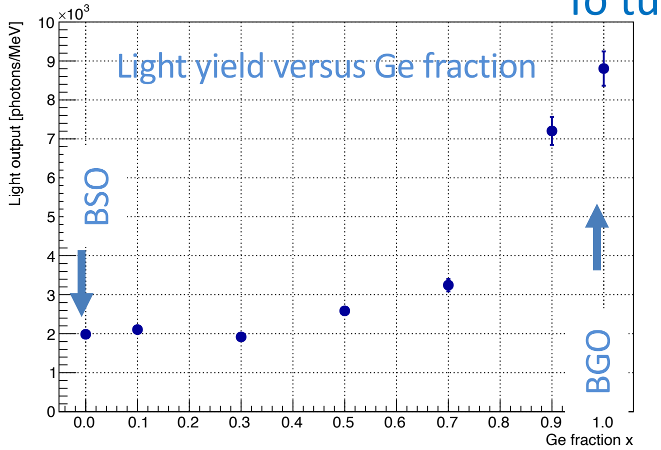


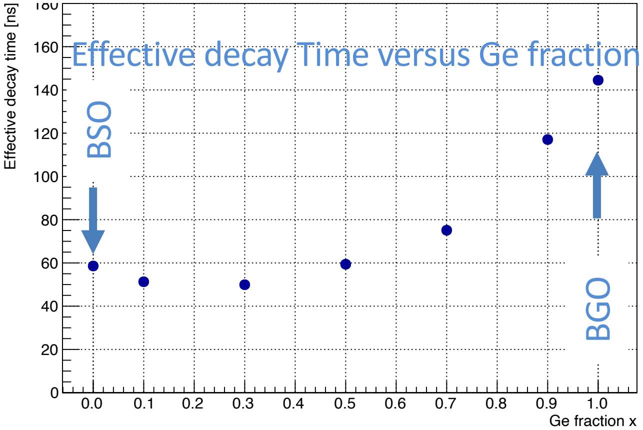
M. Nikl et al, J.Cryst. Growth **229**, 312-315 (2001)
M. Nikl, et al, Radiation Measurements **33**, 705-708 (2001)
M. Kobayashi, et al: Nucl. Instr. Meth. in Phys. Res. A **459**, 482-493 (2001)

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

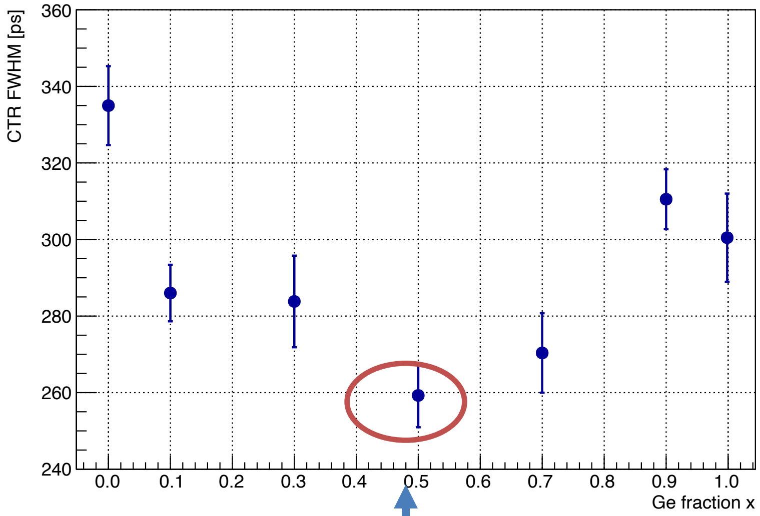
Mixed Material: BGO-BSO ($Bi_4(Ge_xSi_{1-x})_3O_{12}$)

To tune the material properties





Coincidence time resolution @511Kev versus Ge fraction



BSO

BGO

Optimal Ge fraction for time resolution

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

Exploiting Cerenkov Emission

3 types of materials:

- **Pure Cerenkov** as PbF_2, undoped heavy materials (eg LuAG):

"A Cerenkov EM-calorimeter for CMS, using PbF_2 crytals" proposed by J. L. Faure, 1992
Currently under study for Klever/Crylin calorimeter

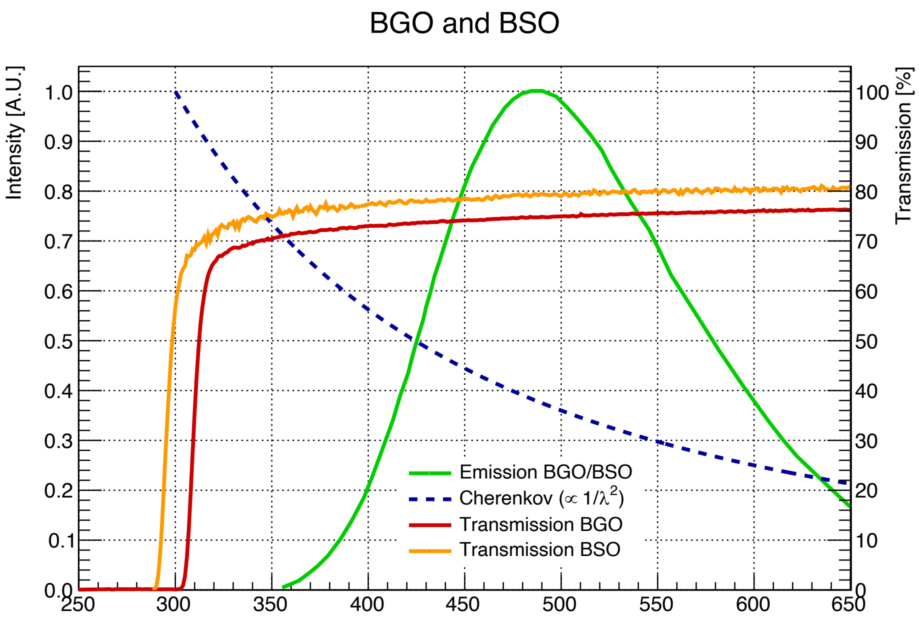
- **Cerenkov+semiconductor** as TlBr, TlCl

- **Cerenkov + Scintillation:**
  - heavy scintillator: eg PWO, BGO, BSO, LuAG:Ce, Pr
  - Light scintillator: Silica doped materials

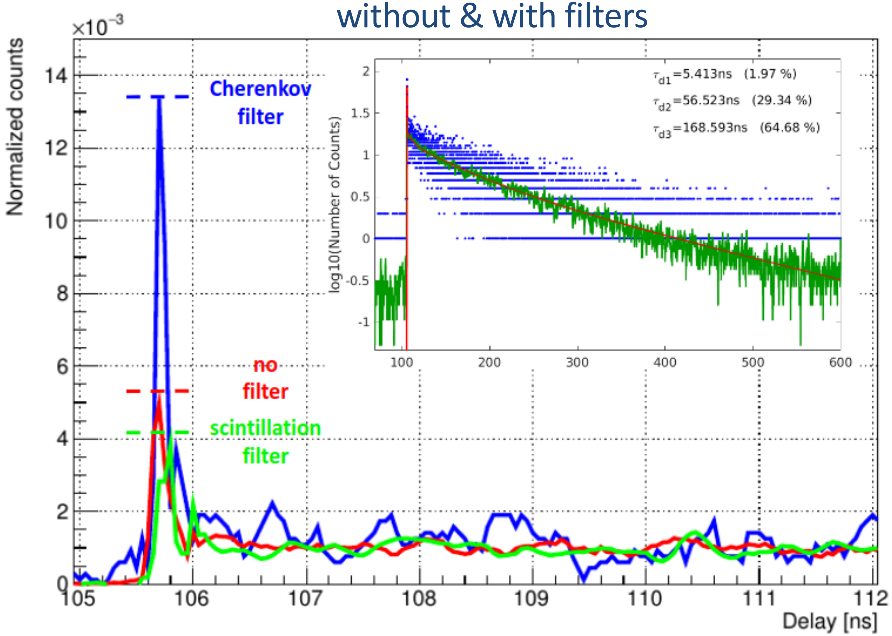
  => dual readout with same material by separation emission wavelength or pulse shape

See talks Marco, Romualdo today

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 and/or pulse discrimination

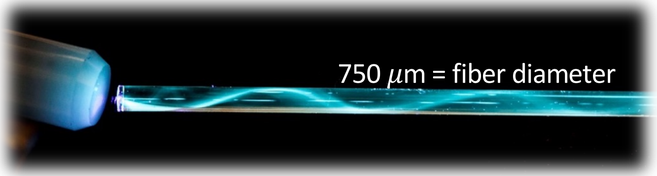
BSO (or mixed BGSO) is faster than BGO and has higher LY than PWO

⇒ Promising candidate for dual readout homogenous calorimeter

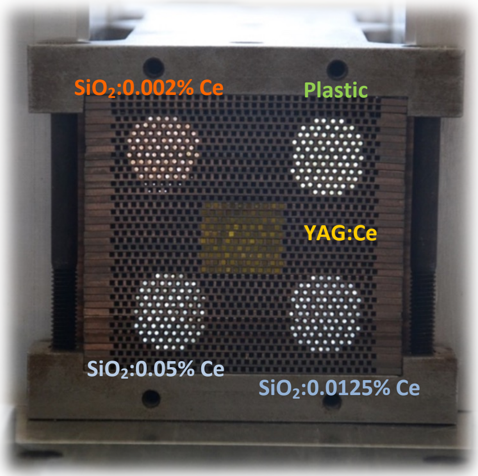
R. Cala et al, NIMA 1032, 2022, 166527

Exploitation of Cerenkov/scintillation in Silica doped fibers

SiO2:Ce **fibers** Milano/Polymicro

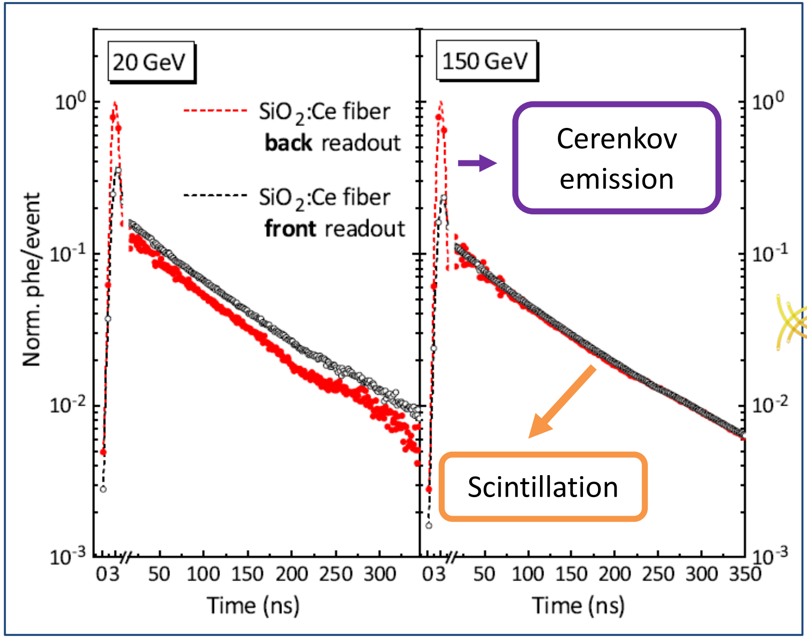


Test with 20GeV in CERN SPS



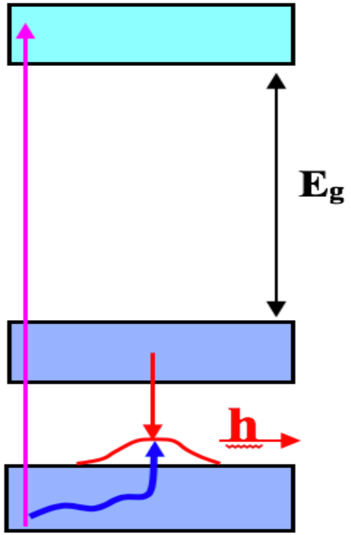
F. Cova et al., Phys. Rev. Appl. 11 (2), 024036 (2019)

Dual read-out of Cerenkov and scintillation light simultaneously with the same SiO_2:Ce fiber



Crossluminescence Materials

Radiative transition between the core- and valence bands.



Very fast emission < 2ns
but generally emission < 300nm

Many possible materials:

Compilation of CL data at 293 K

C.W.E. Van Eijk Journal of Luminescence60, 1994, 936-841

	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]

BaF_2 →

BaF_2 was proposed in 90's for ECAL for L* @SSC and L3P @LHC

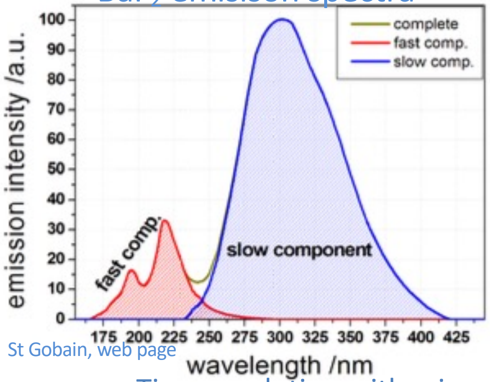
L* Collaboration, Letter of Intent to the SSC Laboratory: https://lss.fnal.gov/archive/other/ssc/sscl-sr-1154.pdf ; R. Zhu, NIMA A 340 (1994) 442-457

Crossluminescence Recent Developments

CRYSTAL CLEAR

R&D to **suppress the slow component by doping**

BaF_2 emisison spectra

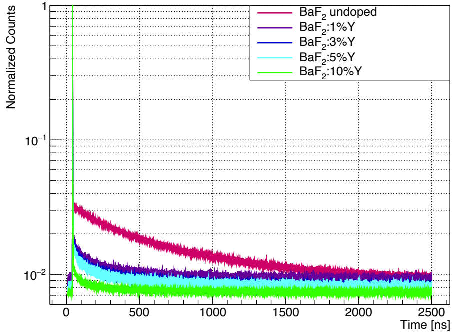


St Gobain, web page

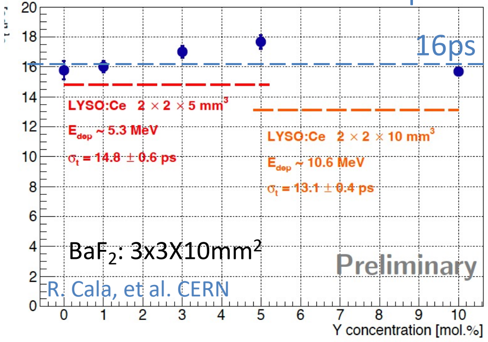
⇒ No change in short decay

⇒ No impact on time resolution

Decay time spectra



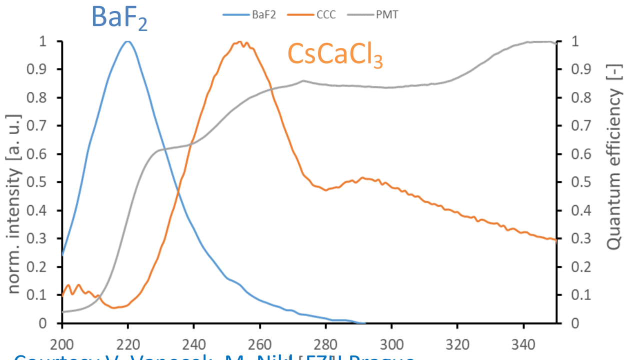
Time resolution with mips



J. Chen, et al., IEEE Trans. Nucl. Sci., vol. 65, no. 8, pp. 2147-2151, 2018.

R. Cala et al, SCINT2022 conference SantaFe Sept2022

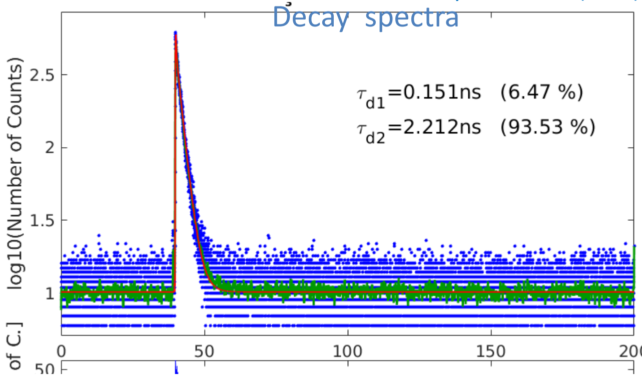
R&D **to shift the emission in UV Visible**



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



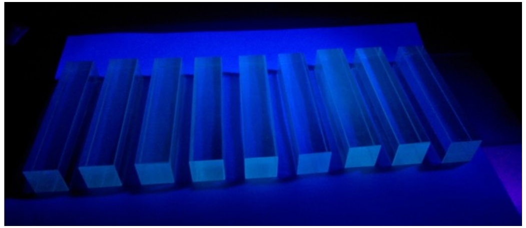
V. Vanecek et al., Optical Materials X 12 (2021) 100103

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

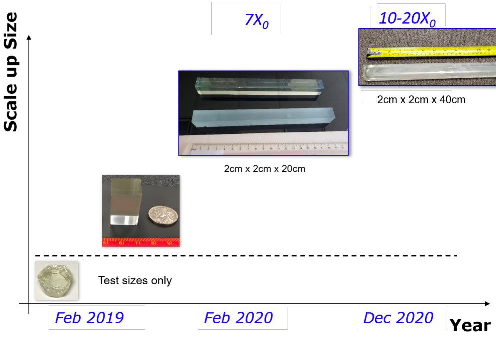


Industrial development via ScintiGlass: Attract project with Preciosa Company

ATTRACT

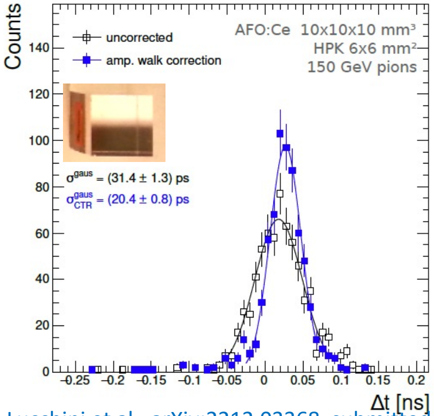
V. Dormenv et al, NIMA, 1015, 2022, 165762

EIC R&D: eRD105 (SciGlass)



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

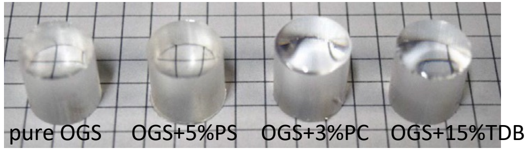
Fluorophosphate AFO glasses Timing resolution with mip

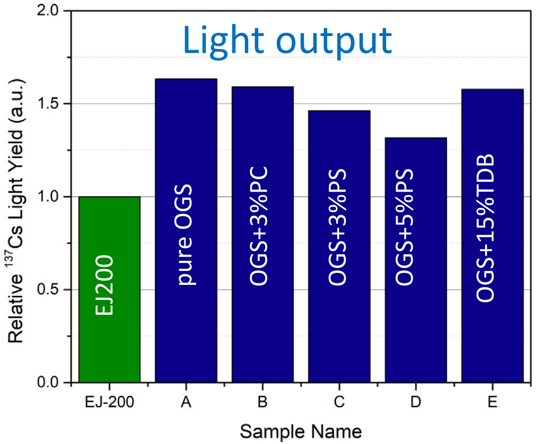


M. Lucchini et al., arXiv:2212.03368, submitted to NIMA

R&D for Organic Scintillators

Organic glasses developed in Sendai National lab



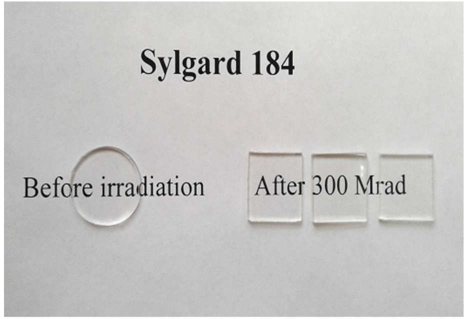


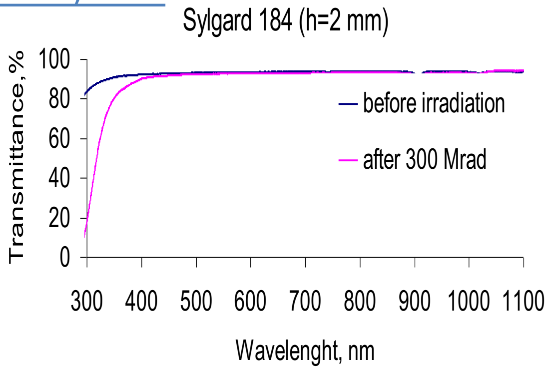
From L. Q Nguyen et al., NIMA 1036 (2022) 166835

Polysiloxane materials

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

ISMA (Kharkiv) tests





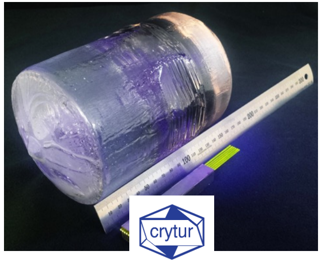
Courtesy A Boyarintsev, ISMA, Kharkiv

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

New Production Methods

Crystal fibre production

Czochralski method
Fibres cut from large ingot



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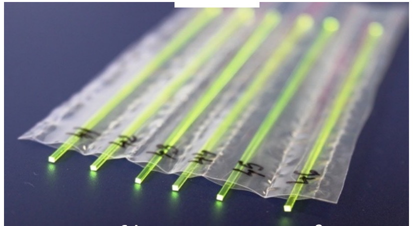
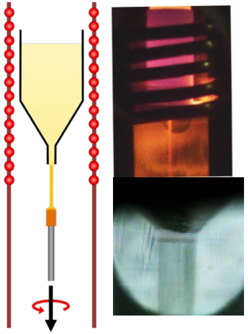


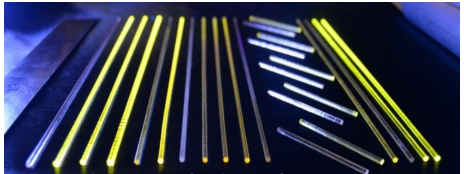
Square fibers 1x1x10mm^3

CRYSTAL CLEAR

Intelum

Micropulling down technique



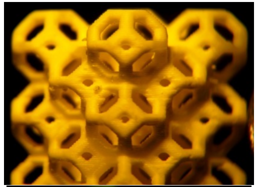


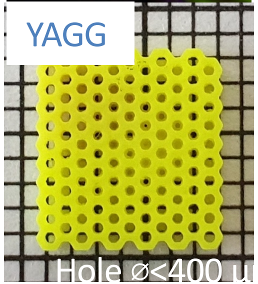
Several types of Fibers from ILM; Lyon

⇒ 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

3D printing of Scintillators

crystals

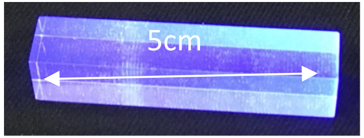


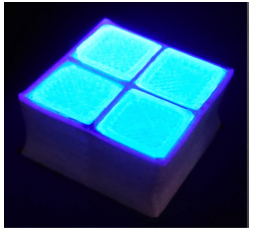


Courtesy of G. Dossovitky, Kurchatov Institute

Plastic scintillator

3D Det project

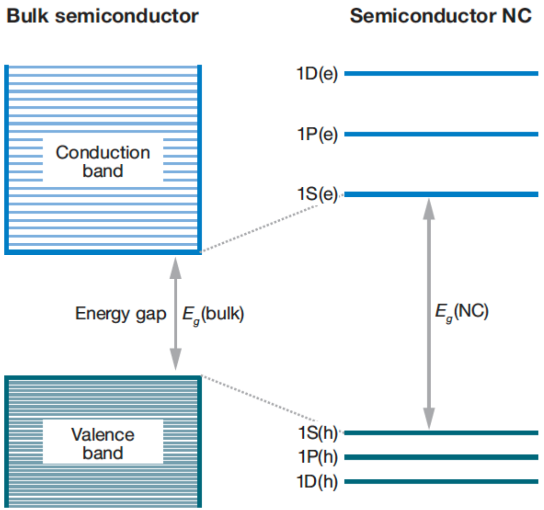




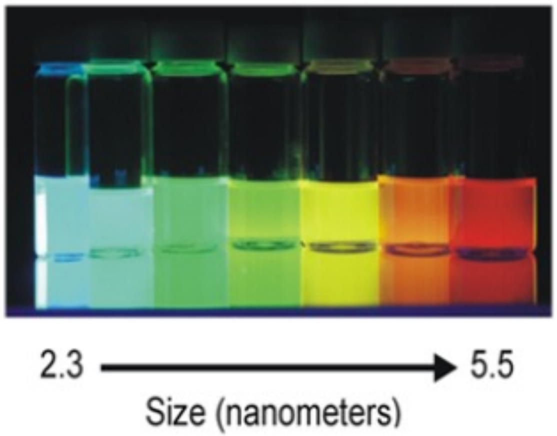
From EP newsletter Nov 21

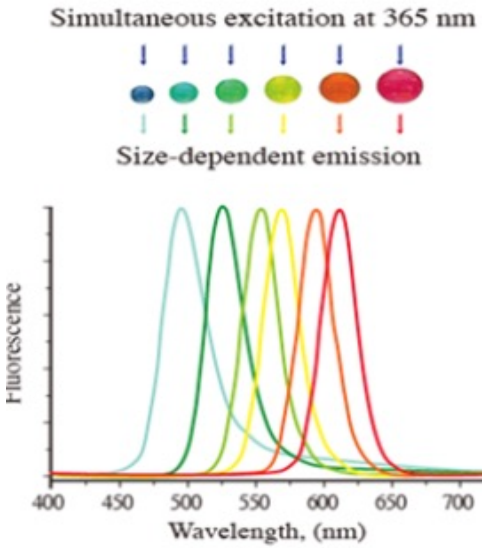
From Bulk to Nanomaterial: Quantum Confinement

Same crystal lattice but nanometer-sized crystal particle



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



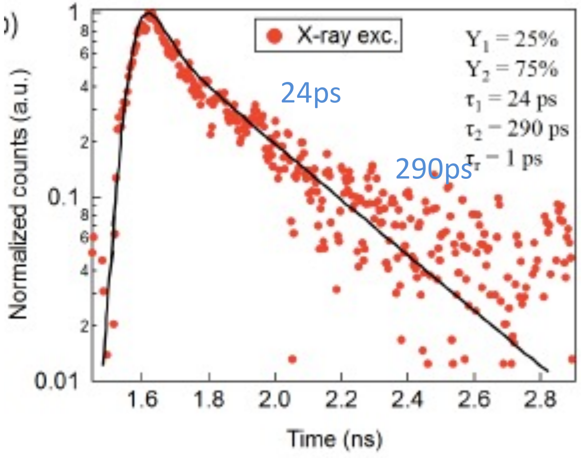


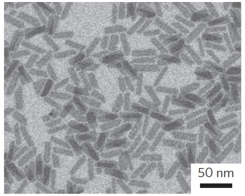
from Benoit Dubertret and Hideki Ooba

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

Some Examples with sub ns Emission

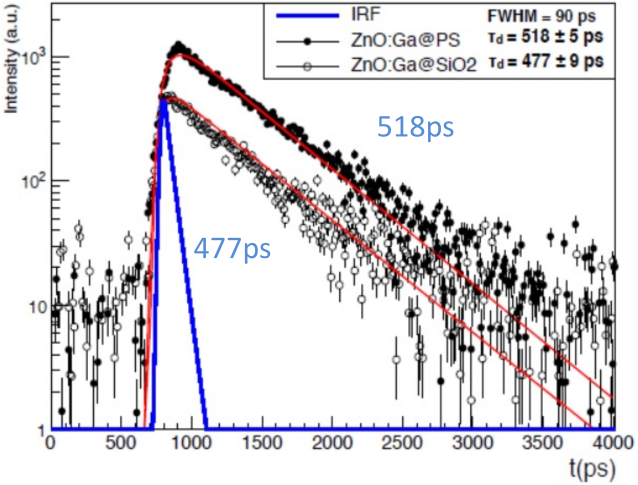
CdSe nanoplatelet,

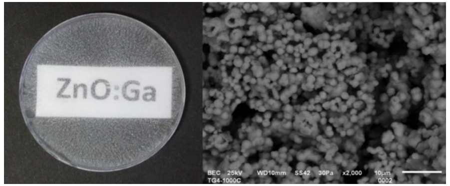




J. Grim et al., *Nature Nanotechnology*, **9**,2014, 891–895
R. Martinez Turtos et al., 2016 JINST_11 (10) P10015

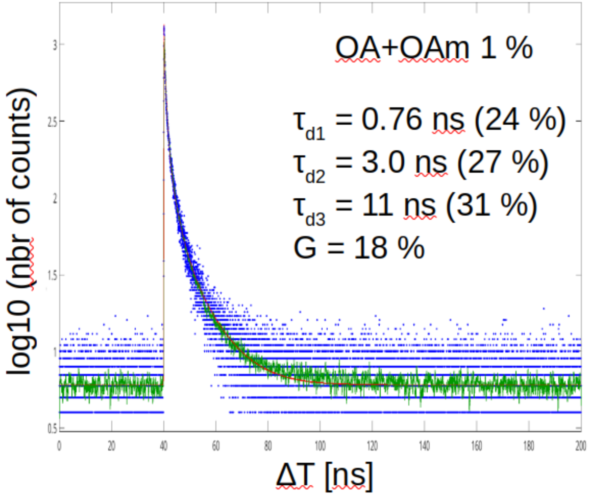
ZnO:Ga embedded in SiO_2 or polystyrene

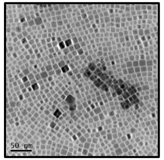




Procházková et al., Radiat Meas 90, 2016, 59-63
R. Turtos Phys. Status Solidi RRL 10, No. 11, 843–847 (2016)

$CsPbBr_3$ embedded in polystyrene

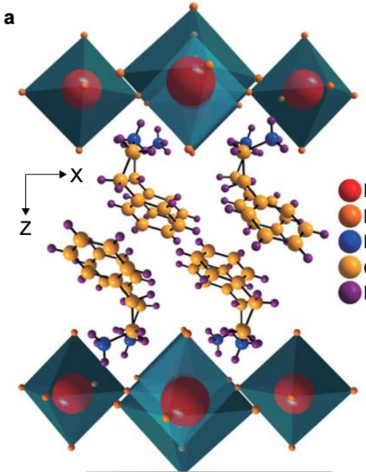


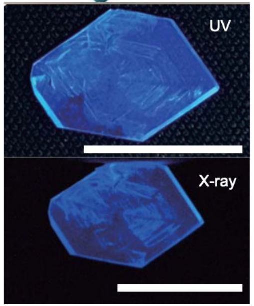


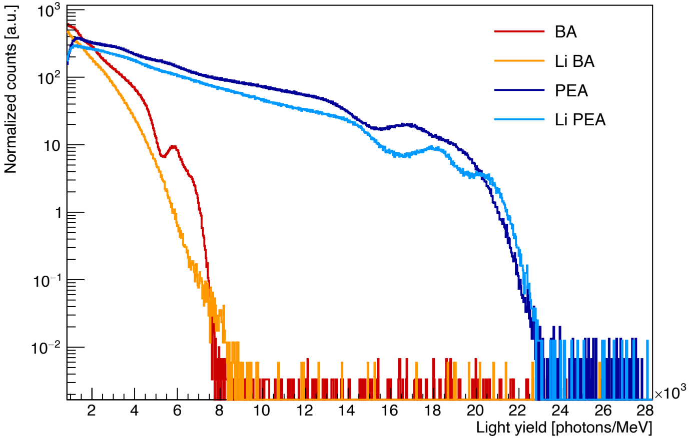
K. Děcká et al, Journal of Material Chemistry C

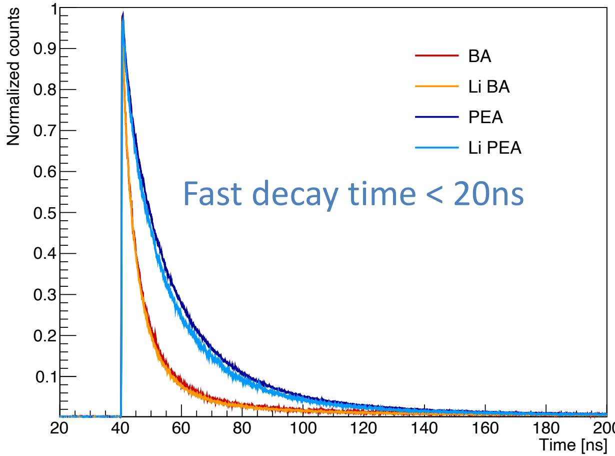
Two-dimensional Hybrid Perovskites

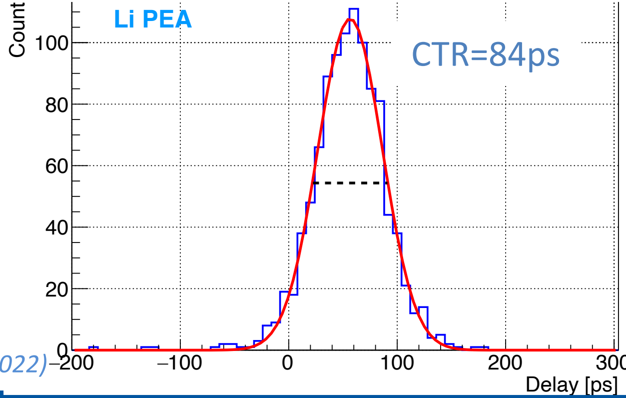
An organic-inorganic hybrid structure.











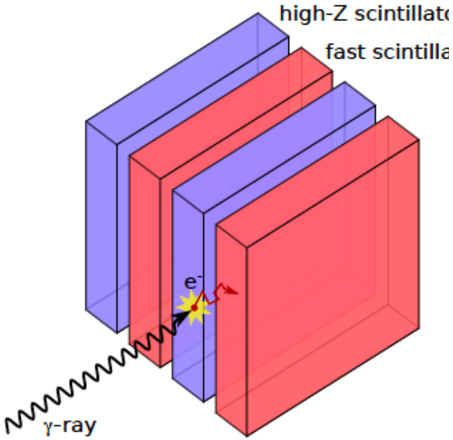
Relatively high light output
20000ph/MeV for PEA type

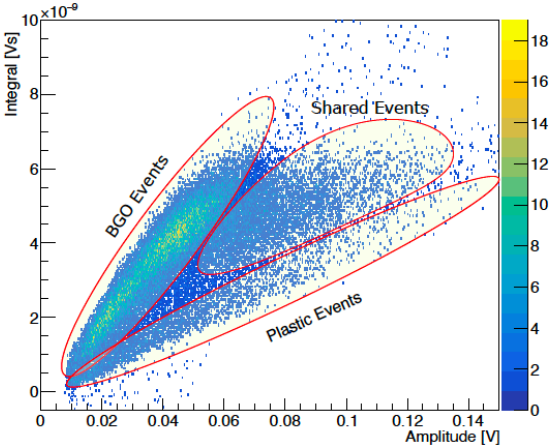
A. Xie et al, Chem. Mater. 2020, 32, 8530–8539

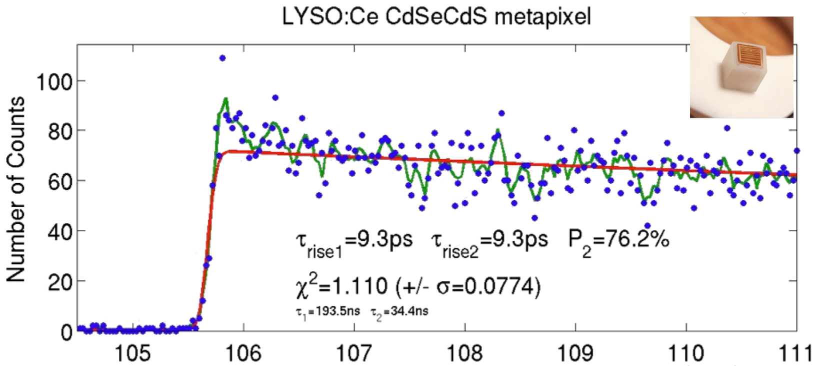
R. Cala et al, Appl. Phys. Lett. 120, 241901 (2022)

Heterostructure Concept

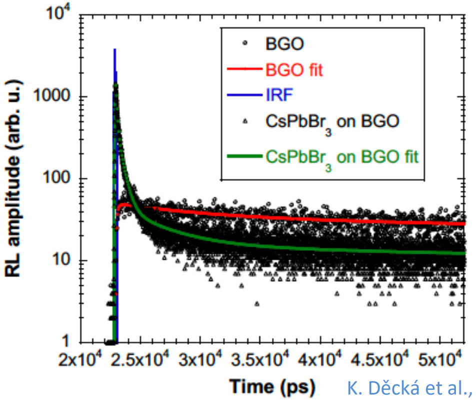
Combine scintillators with high light yield, high stopping power with prompt emission material







R. M. Turtos et al, Phys. Med. Biol. 64 (2019) 85018



BGO

BGO+ $CsPbBr_3$

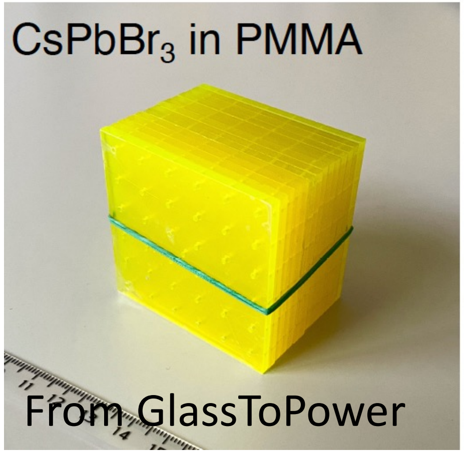
K. Děcká et al., Nanomaterials 2022, 12, 14.

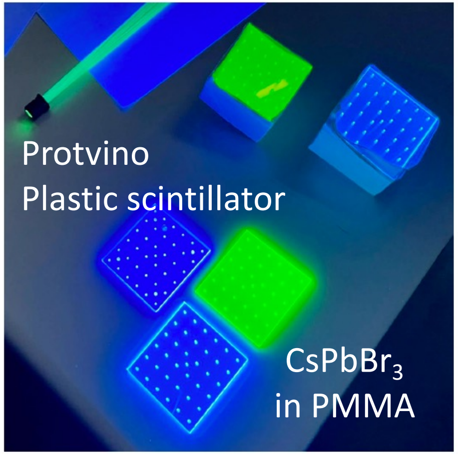
=> Energy sharing between bulk and fast emitter

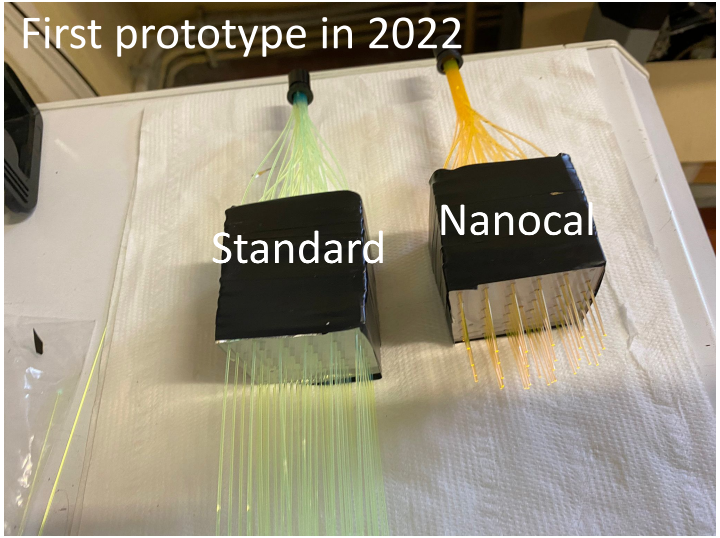
Concept proposed in the frame of ERC TICAL (GA 338953 PI: P.Lecoq)
R. M. Turtos et al, Phys. Med. Biol. 64 (2019) 85018
F. Pagano et al, 2022, 2022 Phys. Med. Biol. 67 135010

First Attempt to use Nanomaterial in HEP
Nanocal Bluesky Aidainnova project

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







First test beam performed in October 2022 in H2

Protvino scintillator	NanoCal scintillator
Polystyrene	PMMA
1.5% PTP/0.04% POPOP	0.2% CsPbBr3
Kuraray Y-11(200) fibers	Kuraray O-2(100) fibers

From M. Moulson Aidainnova WP13 20.12.2022

See EP newsletter Nov 22

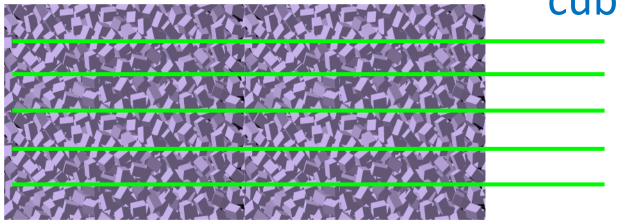
M. Moulson presentation Aidainnova WP13 20.12.2022

Grainita Project

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

Concept: dispersed sub-millimetric grains 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:

$$\frac{\sigma_E}{E} \sim \frac{2\%}{\sqrt{E}}$$

Current Status
- Proof of principle demonstrated with a small test bench
  - (average light path in $ZnWO_4$ + propanol ~ 17 cm)
- Tests of various WLS fibers from Kuraray on-going
- 16-channels prototype (~ 200 g $ZnWO_4$) under design
- Cosmic rays test bench under design

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

R&D over the next 5 years and beyond

For future developments of "optical calorimeters", R&D on scintillator materials is a key item

Several axes of research required depending:

1) on the type of colliders

   High radiation levels/medium/low radiation levels:

2) homogenous/sampling calorimeters

   Dense or light material

R&D over the next 5 years and beyond

R&D on scintillators

- Fast and radiation hard crystals:
  - Engineering of existing materials => Eg. GAGG (LHCb), PWO highly codoped (Crylin, Klever)
  - New composition mixed materials
- Cerenkov/scintillation simultaneous readout => Eg BGO, BGSO, PWO, doped quartz fibers and glasses (IDEA; Calvision)
- Better exploitation of prompt emission
  - Cerenkov, cross-luminescence => Eg. BGO, BGSO, PWO, PbF2, BaF2,.. (timing layers)

  => Requires **fast UV Photo detectors**, UV optical glue, **fast electronics**

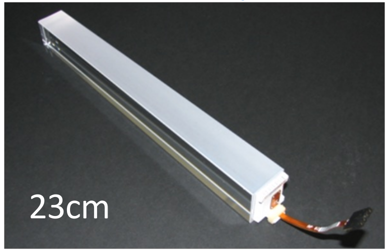
  => Synergy with other DRDs (photodetectors, electronics)
- Glasses, organic material (lower cost materials)
- Innovative materials: nanomaterials, heterostructures materials

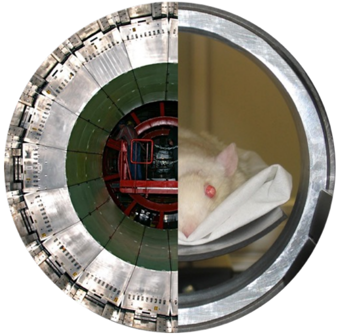
=> R&D on materials required in parallel

  - Development of new characterization and modelisation methods
  - Development of low cost production: work on crucibles, micro-pulling down, 3D printing, etc…
- Exploit synergy with other fields
  - Exploitation of photonic, plasmonic for the enhancement of light extraction, light transport, etc...
- Exploit synergy with other applications

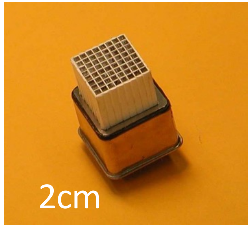
Importance of synergy with other applications
Case of medical applications: PET

ECAL in CMS experiment





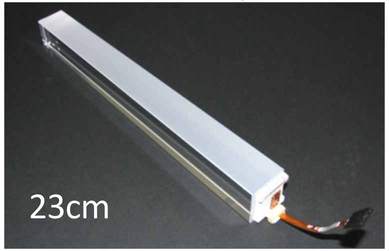
PET scanner

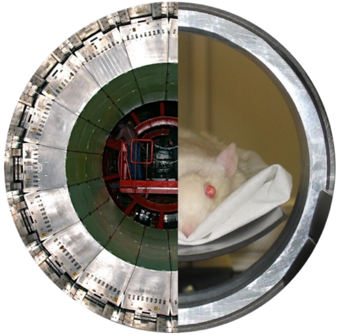


ClearPET module

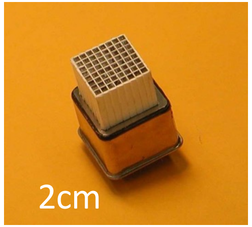
Importance of synergy with other applications
Case of medical applications: PET

ECAL in CMS experiment



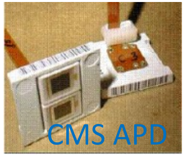


PET scanner



ClearPET module

Example of cross fertilization between HEP to Health:



ClearPEM:
Used APD developed for CMS ECAL

HEP

Health

LYSO CMS BTL

Future CMS barrel timing layer: LYSO/SiPM
and electronic developed first for EndoTOFPET

Conclusion

- Many new emerging technologies and developments exist in the field of scintillators, which open new perspectives for new innovative scintillating detector concepts both for optical homogenous and sampling calorimeters with high granularity & high time resolution

- Scintillator community is very active and already organized through the Crystal Clear Collaboration and the SCINT community
  - provides access to a huge expertise developed over the last 30 years through a wide international network of experts in different fields
  - **DRD on calorimetry** can widely benefit of this existing expertise

- Development on scintillators, photo-detectors, electronics for HEP has impact on many applications
  - $\Rightarrow$ Strong cross fertilization between HEP and applied physics (eg medical and industrial apps).
  - $\Rightarrow$ **DRD on calorimetry** can widely benefit from synergy effects achieved in common R&D projects carried out with research partners active in fields outside HEP

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