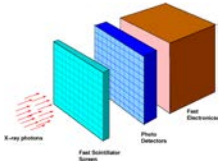


Fast Crystal Scintillators for GHz Hard X-Ray Imaging

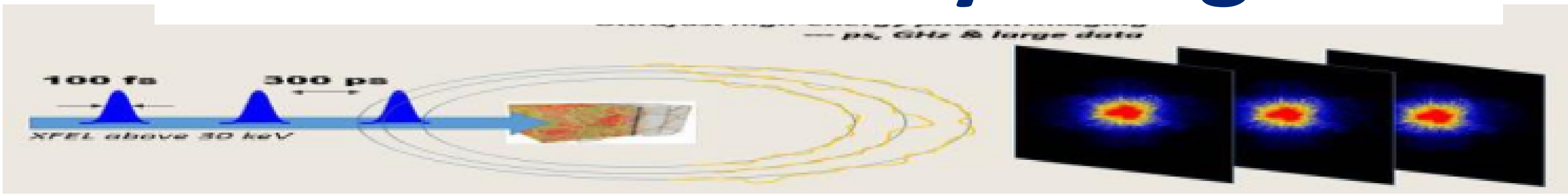
Ren-Yuan Zhu

California Institute of Technology

August 3, 2016

Talk Given at Ultrafast Hard X-Ray Imaging Workshop, Santa Fe, USA

GHz Hard X-Ray Imagers



High-Energy and Ultrafast X-Ray Imaging Technologies and Applications

Organizers: Peter Denes, Sol Gruner, Michael Stevens & Zehui (Jeff) Wang[1]

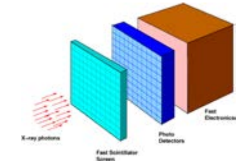
*(**Location/Time**: Santa Fe, NM, USA /Aug 2-3, 2016)*

The goals of this workshop are to gather the leading experts in the related fields, to prioritize tasks for ultrafast hard X-ray imaging detector technology development and applications in the next 5 to 10 years, see Table 1, and to establish the foundations for near-term R&D collaborations.

Table I. High-energy photon imagers for MaRIE XFEL

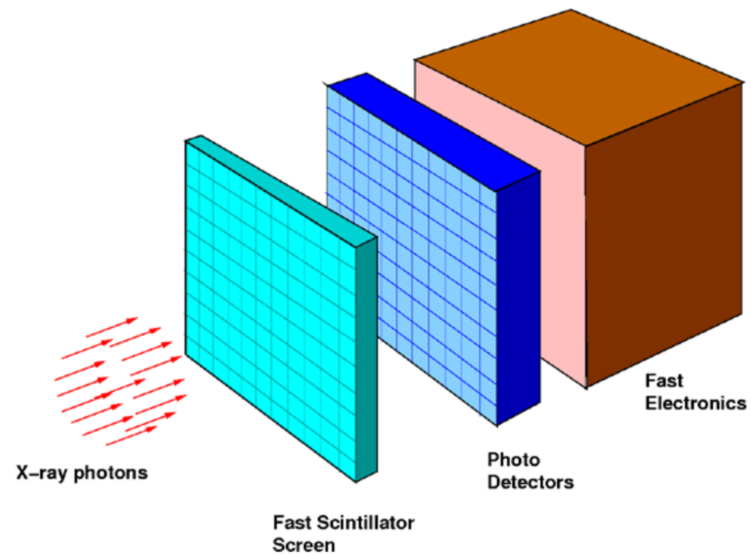
Performance	Type I imager	Type II imager
X-ray energy	30 keV	42-126 keV
Frame-rate/inter-frame time	0.5 GHz/2 ns	3 GHz / 300 ps
Number of frames	10	10 - 30
X-ray detection efficiency	above 50%	above 80%
Pixel size/pitch	≤ 300 μm	< 300 μm
Dynamic range	10^3 X-ray photons	≥ 10^4 X-ray photons
Pixel format	64 x 64 (scalable to 1 Mpix)	1 Mpix

2 ns and 300 ps inter-frame time requires very fast sensor



Why Crystal Scintillator?

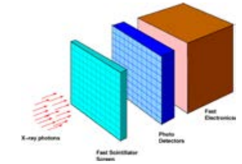
- Detection efficiency for hard X-ray requires bulk detector.
- Fast scintillation light provides fast signal.
- Pixelized crystal detector is a standard for medical industry.
- A detector concept:
  - Pixelized fast scintillator screen;
  - Pixelized fast photodetector;
  - Fast electronics readout.



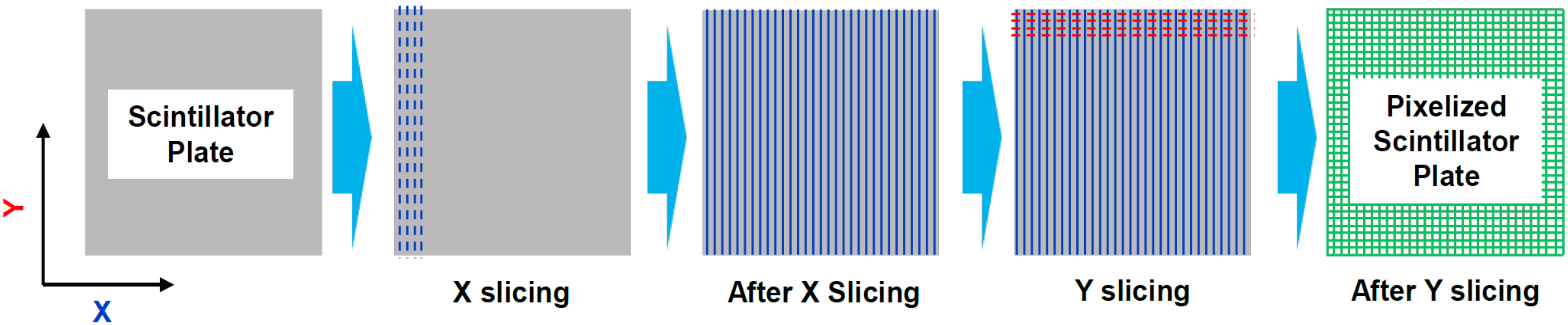
- Challenges:

  Ultra-fast crystals, photodetectors and readout.

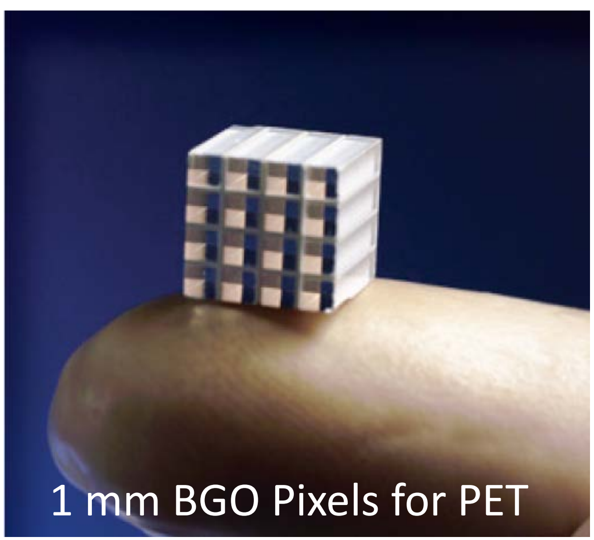
Pixelized Crystal Detectors



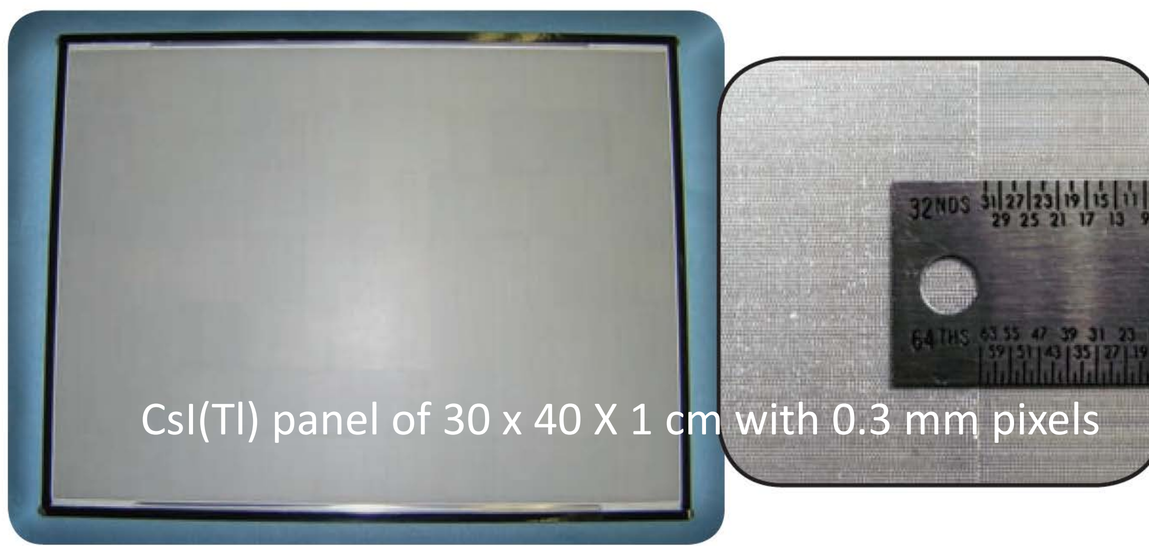
Crystal panels of 300 µ pitch may be fabricated by classical mechanical processing



A Schematic showing pixelized scintillator plate processing



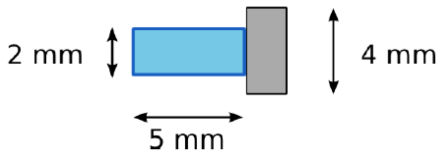
1 mm BGO Pixels for PET



CsI(Tl) panel of 30 x 40 X 1 cm with 0.3 mm pixels

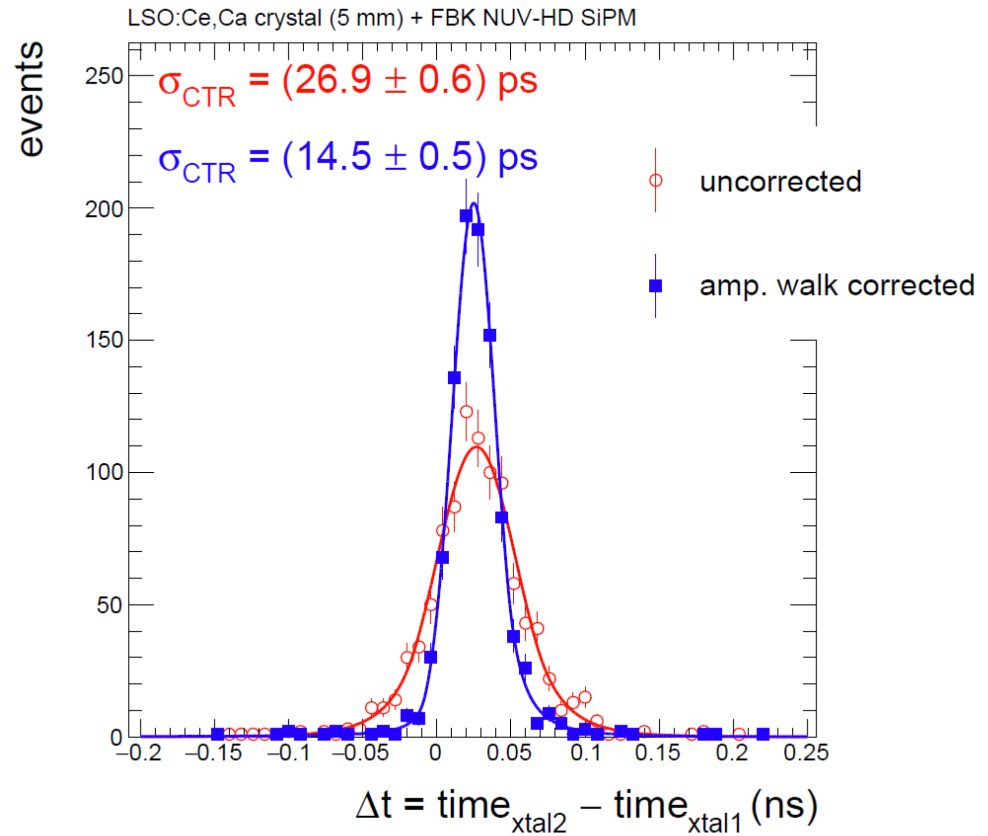
Laser slicing, micropore or not pixelized provide better coverage

Crystals: Excellent Time Resolution



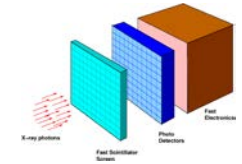
A. Benagia et.al., NIMA 830 (2016) 30-35

- Coincidence time resolution is $\sigma_{\mathbf{CTR}} = 14.5$ **ps** for 5 mm crystals (after correction)
- Assuming that paired crystals have identical response the time resolution of the single device is $\sigma_{\mathbf{single}} = 10.3$ **ps**



LYSO + SiPM provides 10 ps time resolution for MIPs

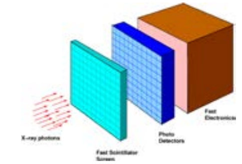
Fast Crystal Scintillators (I)



BaF_2 and wide-gap semiconductor scintillators have sub-ns decay time

	LYSO:Ce	LSO:Ce, Ca[1]	BaF_2	CsF[2, 3]	$CeBr_3$	$LaBr_3$:Ce	YAG:Yb	YAP:Yb	ZnO:Ga	PbI_2[4-6]	GaAs/InAs QDs[7]	Plastic scintillator (BC 404) [8, 9]
Density (g/cm³)	7.4	7.4	4.89	4.12	5.23	5.29	4.56	5.35	5.67	6.16	5.32	1.03
Melting point (°C)	2050	2050	1280	682	722	783	1940	1870	1975	872	1238	70[#]
X_0 (cm)	1.14	1.14	2.03	2.22	1.96	1.88	3.53	2.77	2.51	1.2	2.3	42.54
R_M (cm)	2.07	2.07	3.1	3.76	2.97	2.85	2.76	2.4	2.28	2.76	2.65	9.59
λ_I (cm)	20.9	20.9	30.7	38.9	31.5	30.4	25.2	22.4	22.2	30.6	27.4	78.8
Z_{eff}	64.8	64.8	51.6	52.6	45.6	45.6	30	31.9	27.7	69	32.1	5.82
dE/dX (MeV/cm)	9.55	9.55	6.52	5.32	6.65	6.9	7.01	8.05	8.42	7.29	7.33	2.02
λ_{Peak}[a] (nm)	420	420	300 220	390	371	356	350	350	389	520	1050	408
Refractive Index[b]	1.82	1.82	1.54	1.49	1.9	1.9	1.87	1.96	2.1	3.4	3.47[g]	1.58
Normalized Light Yield[a,c]	100	116[h]	42 4.8	6.6	99	153	0.36[d]	0.19[d]	6.6[d]	133[d,e]	800	35
Total Light Yield (photons/MeV)	30,000	34,800[h]	13,000	2,000	30,000	46,000	110[d]	57[d]	2,000[d]	40,000[d,e]	240,000	11,000
Decay Time[a] (ns)	40	31[h]	650 0.9	3	17	20	4	1.5	<1	1.4[f] 0.29[f] 0.06[f]	1	1.8
Light Yield in 1st ns (photons/MeV)	740	952[h]	960	560	1,700	2,200	24[d]	28[d]	610[d]	4,000[d,e]	150,000	4,500
40 keV Att. Length (1/e, mm)	0.185	0.185	0.106	0.116	0.277	0.131	0.439	0.314	0.407	0.087	0.288	44.2
Issues			Slow component	hygroscopic	hygroscopic	hygroscopic	Self-absorption	Self-absorption	Self-absorption	Self-absorption	Concept	Low detection efficiency

Fast Crystal Scintillators (II)



Notes:

a. Top line: slow component, bottom line: fast component;

b. At the wavelength of the emission maximum;

c. Excited by Gamma rays;

d. Excited by Alpha particles;

e. At 14 K;

f. At 165 K;

g. At 1064 nm;

h. For 0.4 at% Ca co-doping

#. Softening point.

References:

[1] Merry A. Spurrier, et al., *IEEE TNS*,55 (3) 2008: 1178-1182.

[2] M. Moszvnski et al, *NIM A* 205 (1983) 239-249;

[3] http://scintillator.lbl.gov

[4] S E Derenzo et al, *NIM A,* 486 (2002) 214-219;

[5] S E Derenzo et al, *J. Lumin,* 134(2013) 28-34;

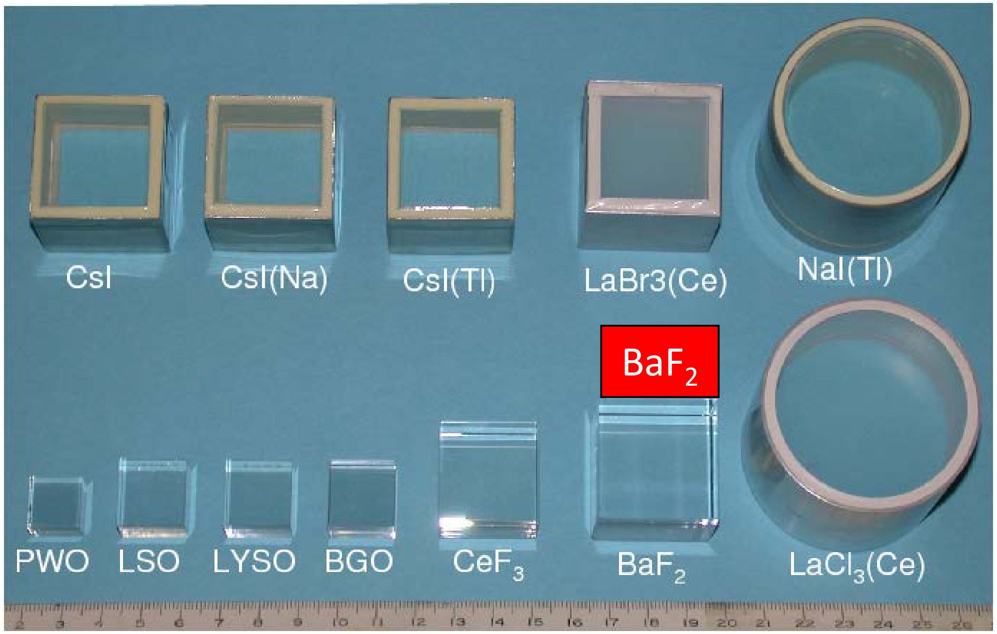
[6] A E Duguan et al, *J. Phyr. Chem. Solids,* 28(1967) 971

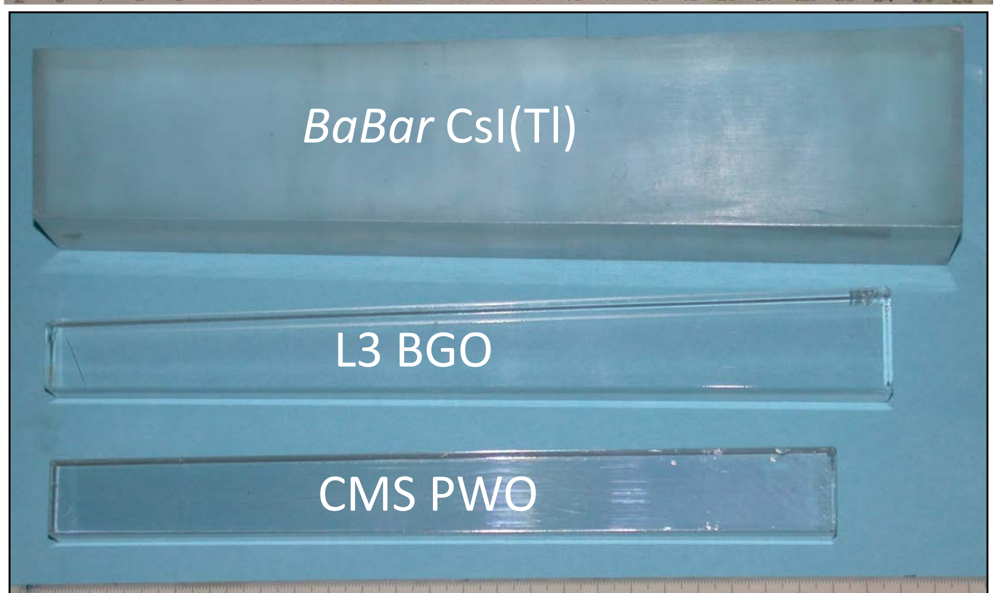
[7] S. Oktyabrsky, et al., *IEEE Trans. Nucl. Sci*. 63, 656 (2016).

[8] http://pdg.lbl.gov/2008/AtomicNuclearProperties/HTML_PAGES/216.html

[9] http://www.detectors.saint-gobain.com/Plastic-Scintillator.aspx;

Crystal Choice





1.5 X_0 Samples:

Hygroscopic: Sealed

Surfaces: Polished

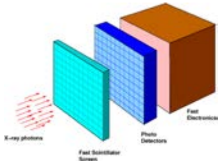
Full Size Crystals:

BaBar CsI(Tl): 16 X_0

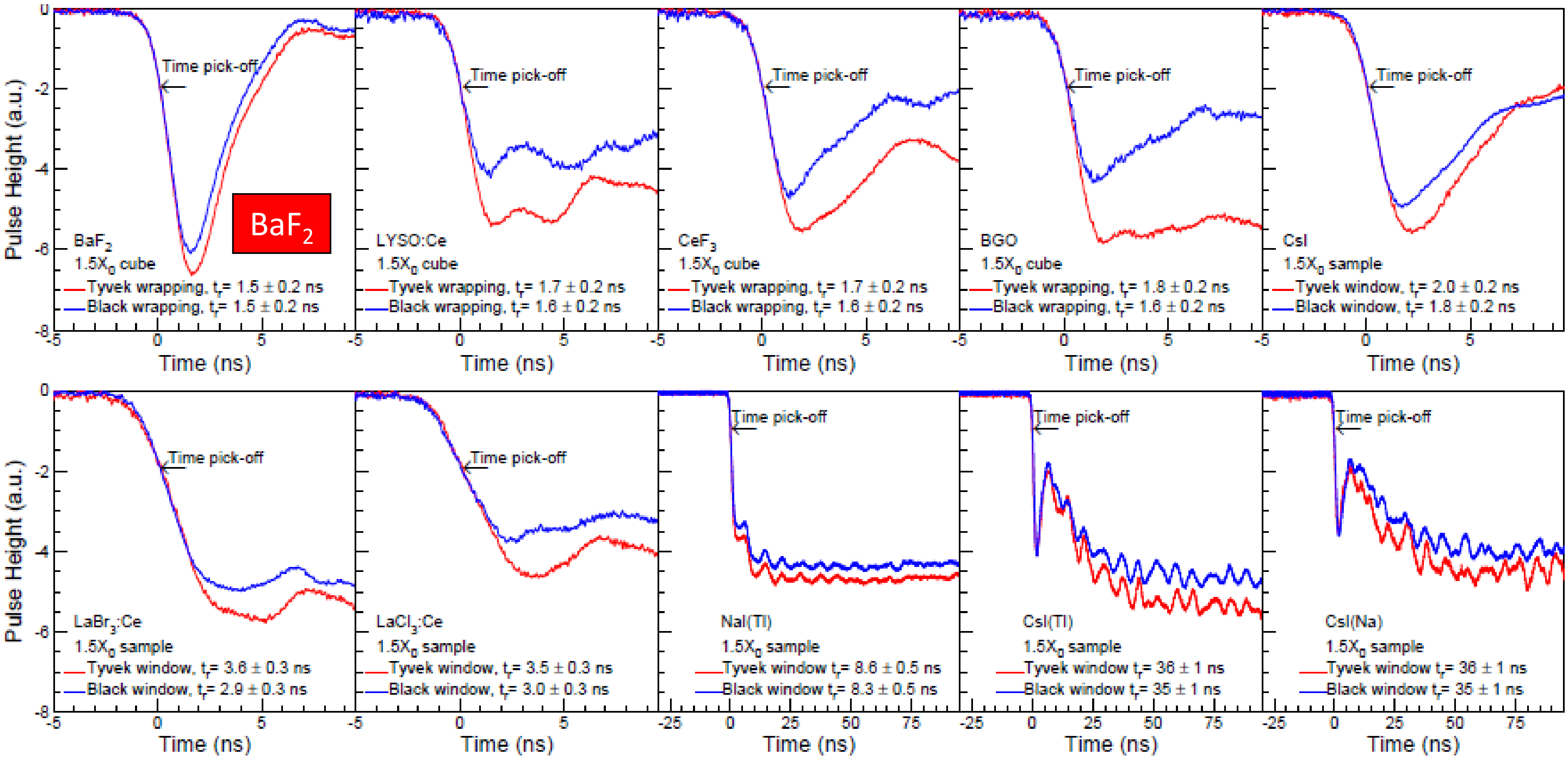
L3 BGO: 22 X_0

CMS PWO(Y): 25 X_0

Fast Signals: 1.5 X_0 Samples



Hamamatsu R2059 PMT (2500 V)/Agilent MSO9254A (2.5 GHz) DSO with 1.3/0.14 ns rise time

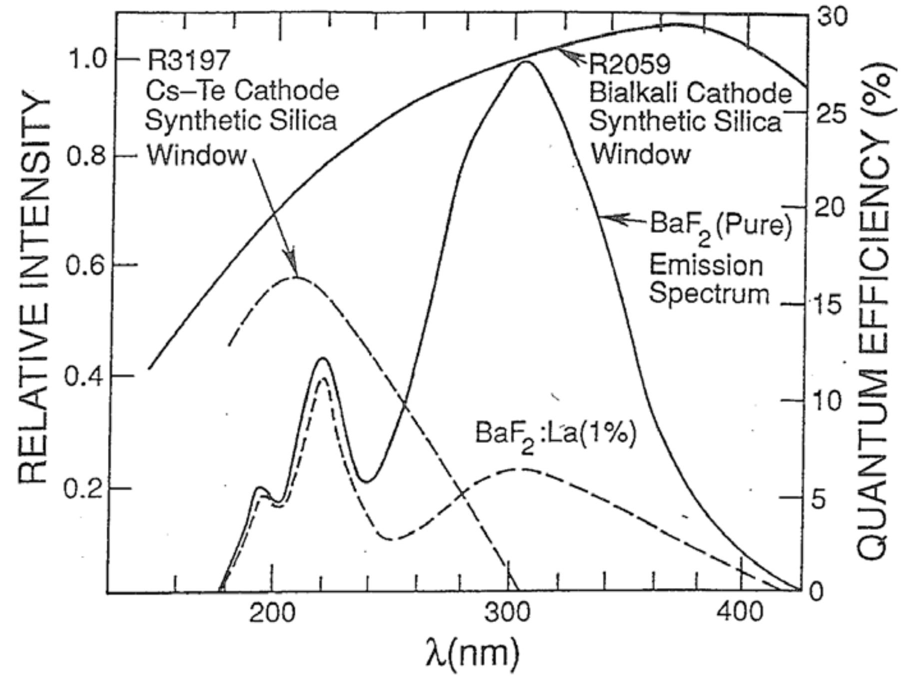


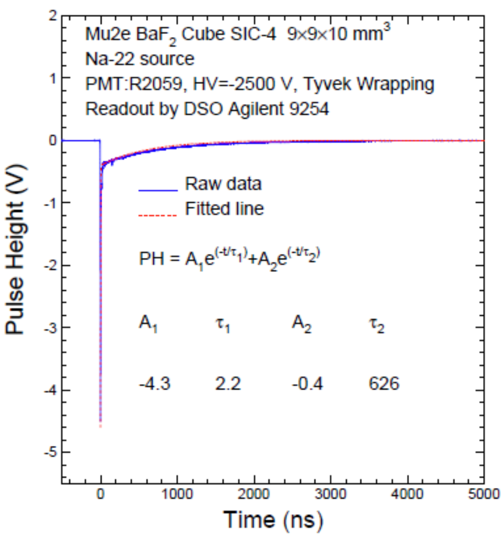
BaF_2 provides narrow pulse width, which will be reduced for small pixels
LYSO & $LaBr_3$ have tail after 2/0.3 ns, causing pile-up for the next frame

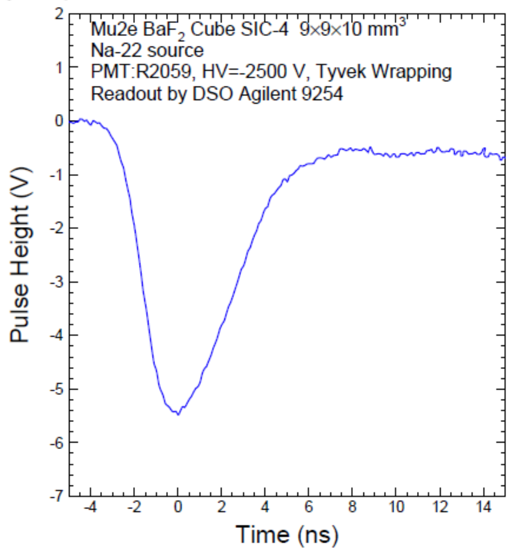
Fast and Slow Signal from BaF_2

The Light output of the fast component of BaF_2 crystals at 220 nm with 0.6 ns decay time is similar to pure CsI.

Spectroscopic selection of fast component may be achieved with solar blind photocathode and/or selective doping.



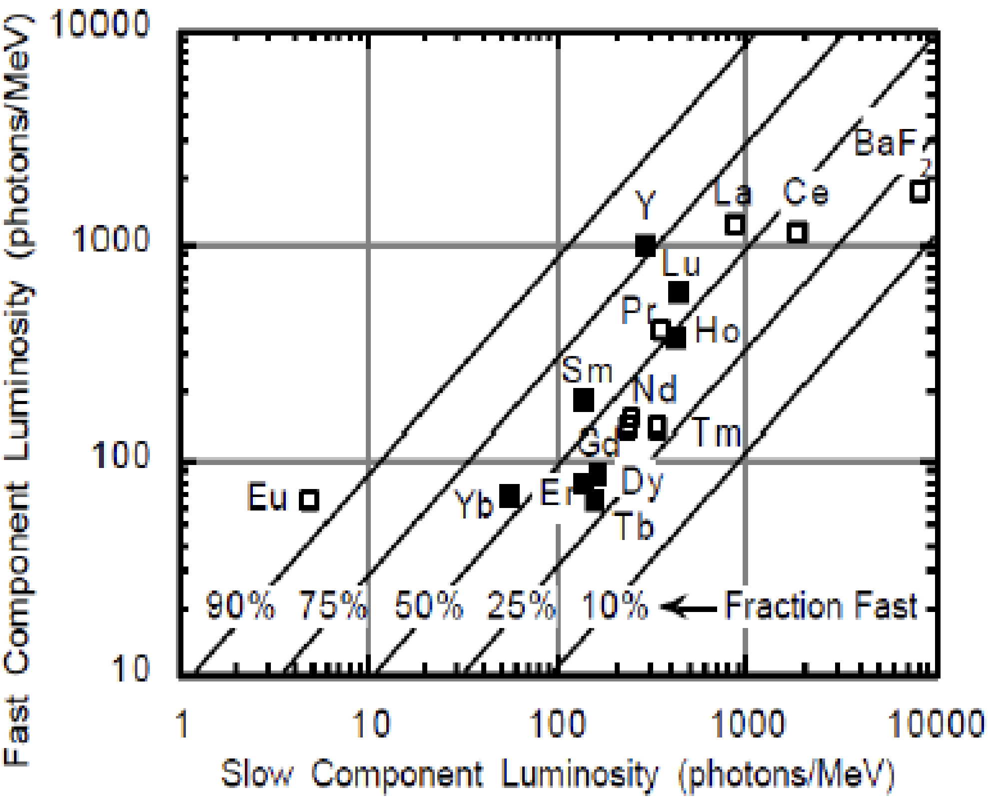


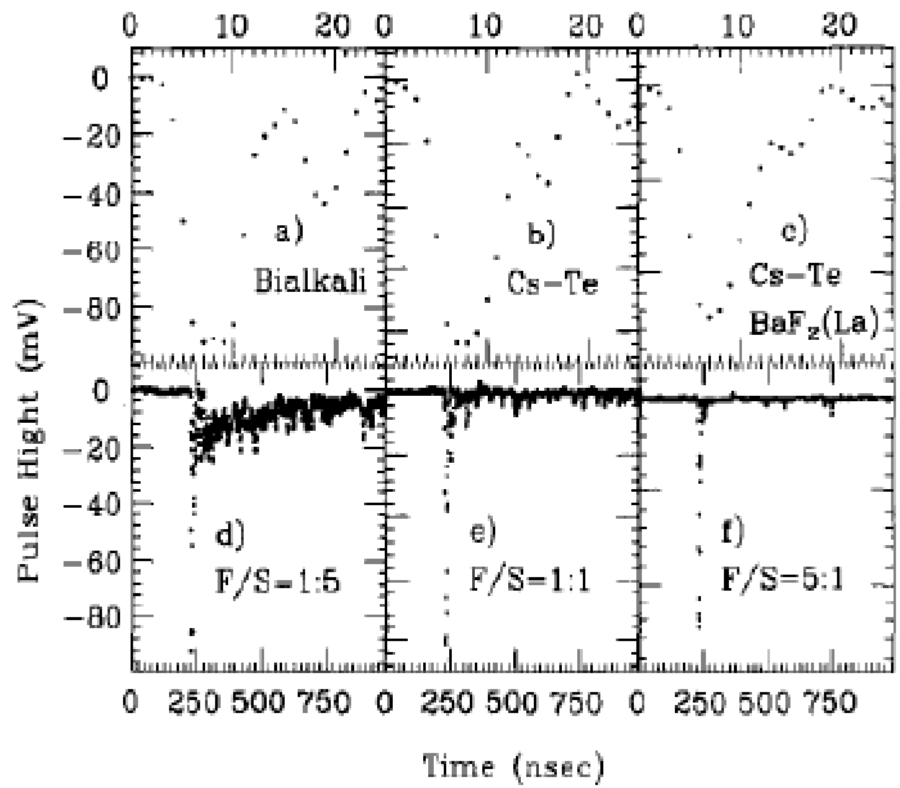


Slow Suppression: Doping & Readout

A very fast BaF_2 calorimeter is proposed for future HEP calorimeter

B.P. SOBOLEV et al., "SUPPRESSION OF BaF2 SLOW COMPONENT OF X-RAY LUMINESCENCE IN NON-STOICHIOMETRIC Ba0.9R0.1F2 CRYSTALS (R=RARE EARTH ELEMENT)," *Proceedings of The Material Research Society: Scintillator and Phosphor Materials*, pp. 277-283, 1994.

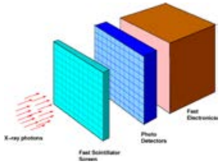




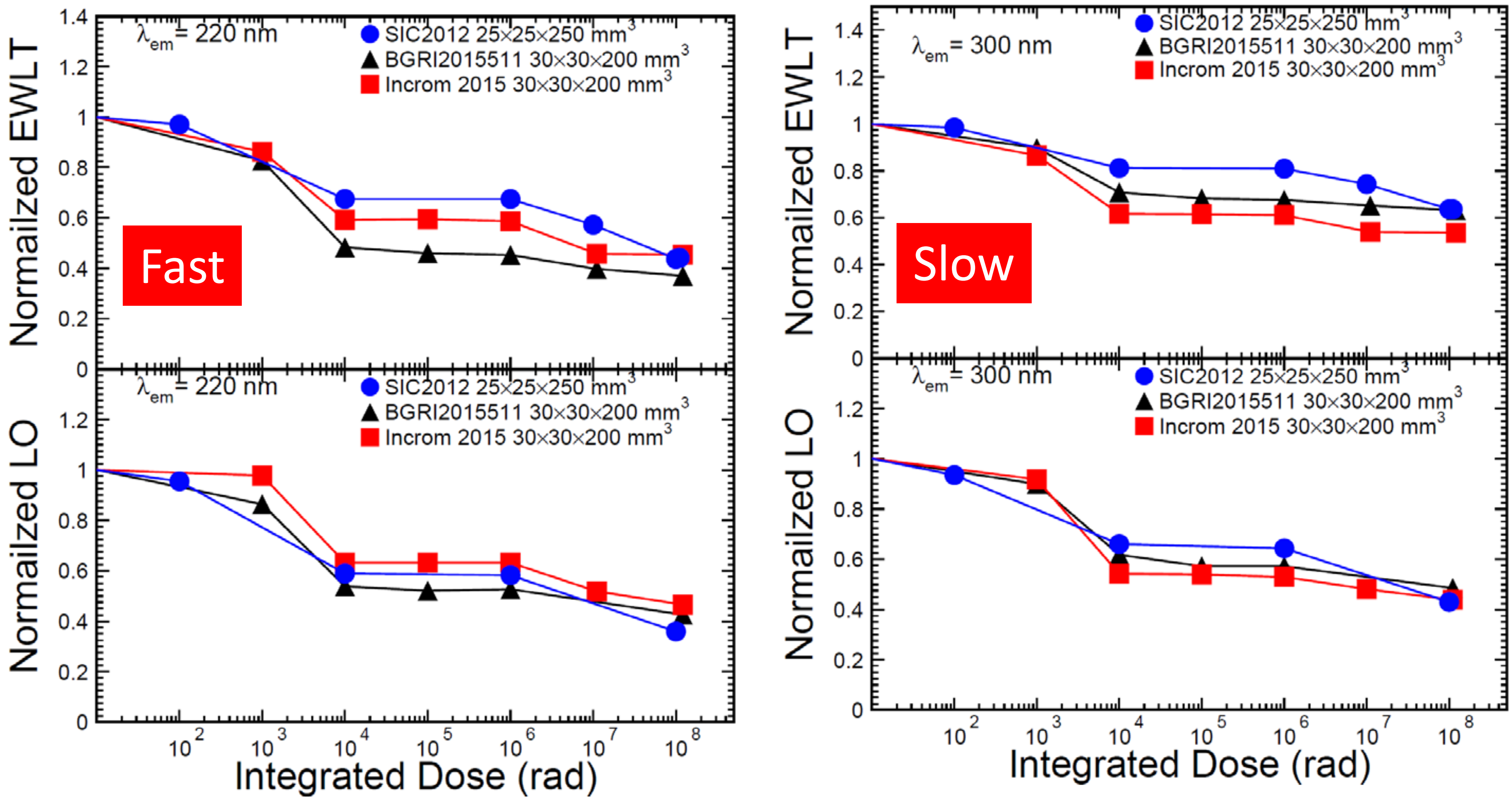
R&D for BaF_2 with slow suppression and solar-blind photodetectors

Z. Y. Wei, R. Y. Zhu, H. Newman, and Z. W. Yin, "Light Yield and Surface-Treatment of Barium Fluoride-Crystals," *Nucl Instrum Meth B*, vol. 61, pp. 61-66, Jul 1991.

BaF_2 is Radiation Hard

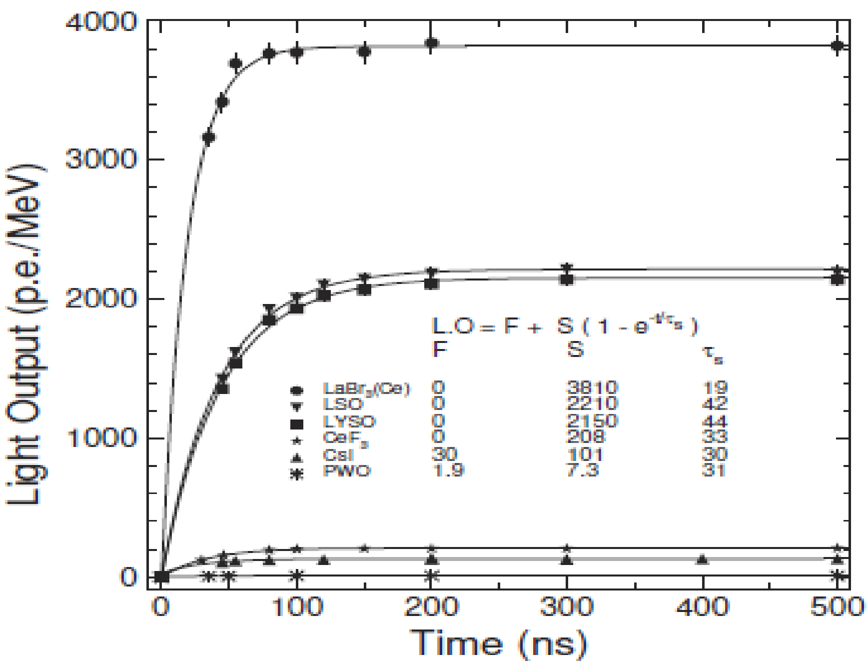


Consistent damage in crystals from three vendors



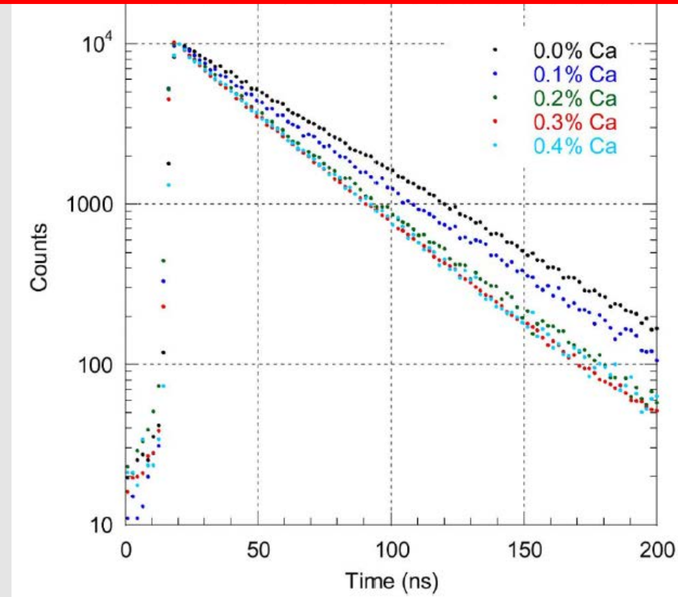
LO after 120 Mrad: 40%/45% for the fast/slow in 20 cm BaF_2, no effect for 0.2 mm

LYSO:Ce and LSO:Ce,Ca Crystals



Light output measured by using a XP2254b PMT is shown as a function of integration time for six fast crystal scintillators[1].

Ca^{2+} co-doping increase the light output in 1^{st} ns



Scintillation decay of LSO:1at%Ce crystals with 0, 0.1, 0.2, 0.3, and 0.4% Ca doping[1]

Properties of LSO:0.1at%Ce,Ca crystals[1]

Ca concentration (%)	Light output (photons/MeV)	Decay time (ns)
0.0	30900	43.0
0.1	38800	36.7
0.2	36200	33.3
0.3	32400	31.3
0.4	34800	31.0

Light output and decay time values are the average of multiple samples.

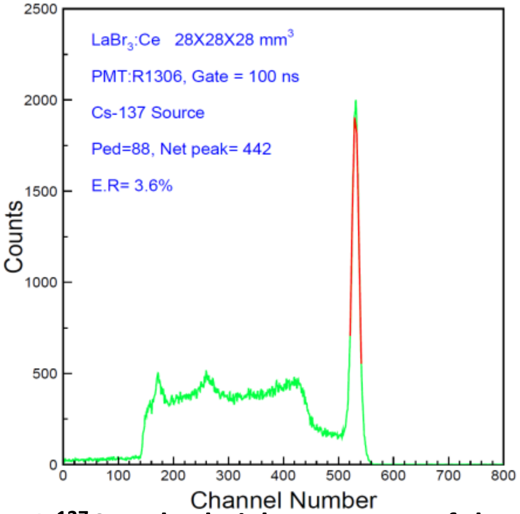
References.:

[1] R. Y. Zhu, Phys. Proc, 37 2012: 372-383.

[2] Merry A. Spurrier, et al., IEEE TNS,55 (3) 2008: 1178-1182.

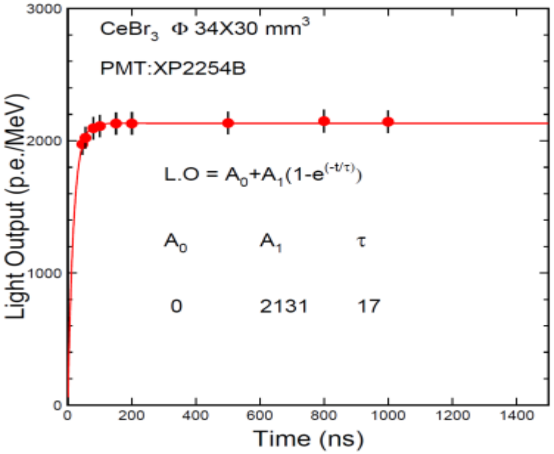
$LaBr_3$:Ce and $CeBr_3$

A photograph of a sealed $LaBr_3$:Ce crystal sample from Saint-Gobain.

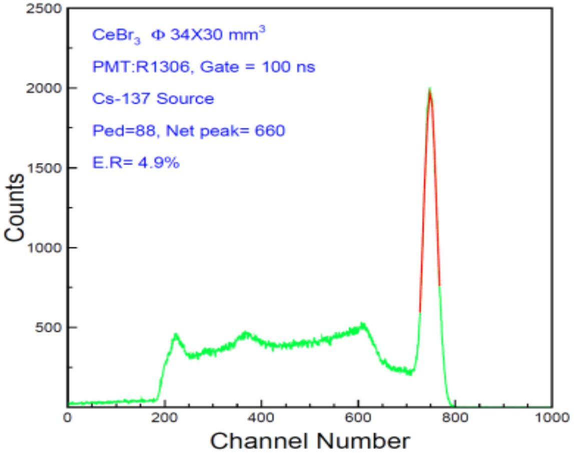


A ^{137}Cs pulse height spectrum of the $LaBr_3$:Ce crystal sample from Saint-Gobain.

A sealed $CeBr_3$ crystal sample from Scionix Holland.

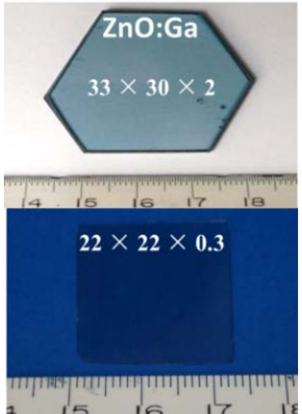


Decay Kinetics of the $CeBr_3$ sample.

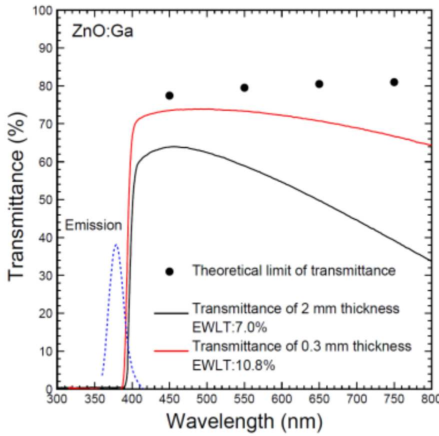


A ^{137}Cs pulse height spectrum of the $CeBr_3$ crystal.

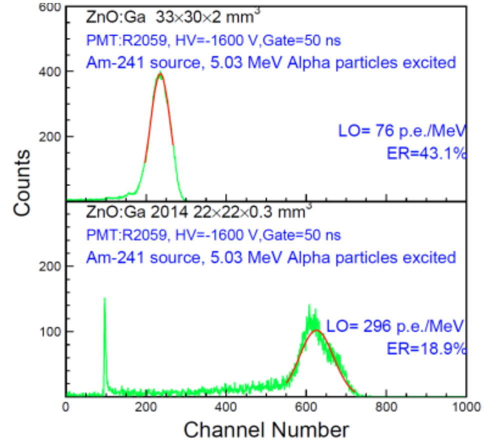
Wide-gap Crystals: ZnO:Ga and PbI_2



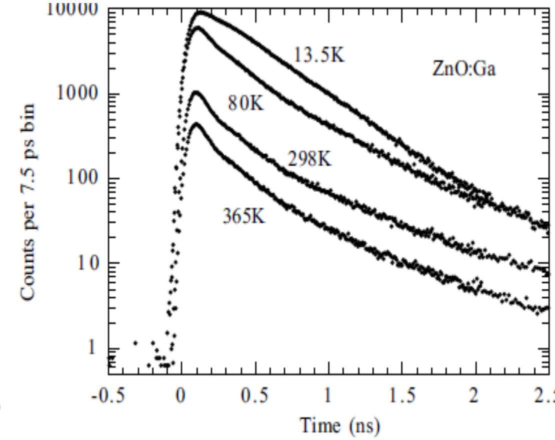
Two ZnO:Ga samples of 2 mm and 0.3 mm thick grown at FJIRSM.



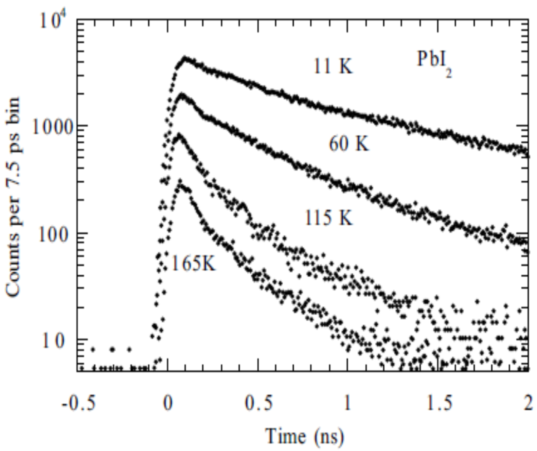
Transmittance and emission spectra of two ZnO:Ga samples of 2mm and 0.3 mm thick.



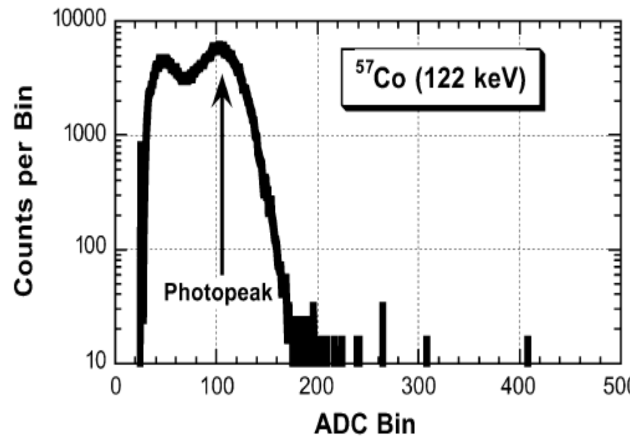
PHS of 5.03 MeV α particles in ZnO:Ga samples of 2 mm and 0.3 mm thick.



Decay Kinetics of ZnO:Ga at room temperature[1].



Decay Kinetics of PbI_2 at different temperature[1].



Pulse height spectra of a Φ 1 x 1 mm PbI_2 sample under ^{57}Co gamma-rays excitation at 8.5 K[2].

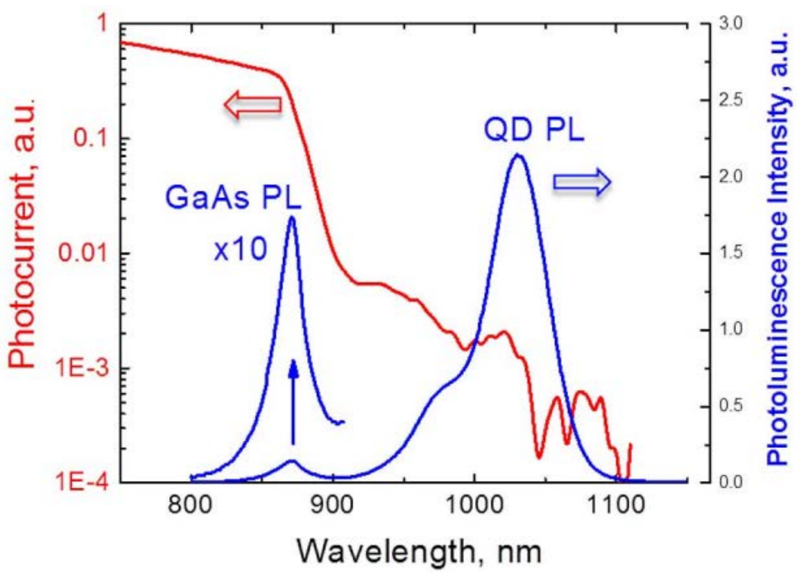
References.:

[1] S. E. Derenzo, M. J. Weber, and M. K. Klintenberg, "Temperature dependence of the fast, near-band-edge scintillation from CuI, HgI_2, PbI_2, ZnO : Ga and CdS : In," *Nucl Instrum Meth A,* vol. 486, pp. 214-219, Jun 21 2002.

[2] W. W. Moses, W. S. Choong, S. E. Derenzo, A. D. Bross, R. Dysert, V. V. Rykalin*, et al.*, "Observation of fast scintillation of cryogenic PbI_2 with VLPCs," *IEEE T Nucl Sci,* vol. 51, pp. 2533-2536, Oct 2004.

InAs/GaAs QD based Crystal

In recent study[1], *S. Oktyabrsky, et al. report* an <u>ultrafast, no self-absorption, high-efficient room-temperature semiconductor scintillator</u> based on InAs QDs embedded in a GaAs matrix.



Room temperature photocurrent spectra overlapped with PL spectra of the same QD structure with reduced wetting layer placed in a p-n junction.

Comparison of Some Fast Inorganic Scintillators (source: Scintillator.lbl.gov) With Projected Performance of InAs/GaAs Qd Scintillator

Parameter	BaF_2	LYSO	GaAs/InAs QDs
Density (g/cm^3)	4.89	7.1	5.32
Radiation length, cm	2.03	1.1	2.3
Decay constant, ns	0.8 ns	40	1
Peak emission, nm	195; 220	428	1050
Photon Yield (photons/MeV)	1,400	34,000	240,000
Time between first photons, for 1MeV	0.57ps	1.2 ps	2 fs
Poisson-limited energy resolution at 1MeV (keV) *	62	13	4.8
Radiation hardness, Gy	10^4-10^5	10^4-10^5	>10^4
Coupling efficiency	<50%	<50%	~100%

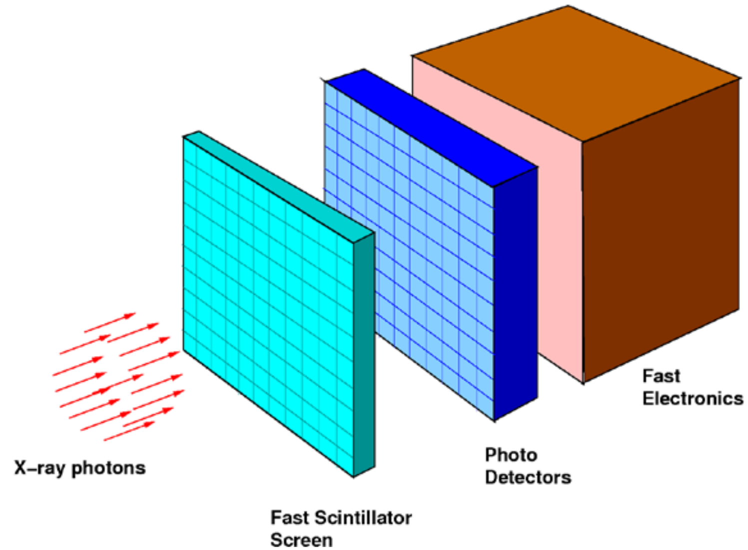
*Assuming collection efficiency = 1

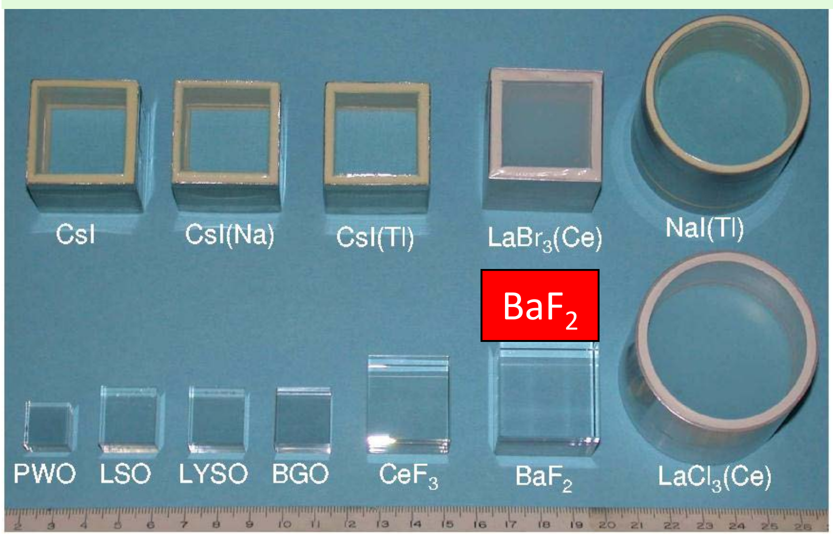
- ☐ **Due to inhomogeneous broadening of very narrow ground-level luminescence peaks, the self-absorption in such a medium is very low, <1 cm^{-1} for 10^{15} QDs/cm^3 [1]**

Ref.: [1] S. Oktyabrsky, et al., IEEE Trans. Nucl. Sci. 63, 656 (2016).

Fast Crystal Scintillators for GHz Hard X-ray Imaging

- ❑ Crystal scintillators with sub-ns decay time, such as BaF_2 may provide fast scintillator based screens for GHz hard X-ray imaging.
- ❑ Wide-band semiconductor based scintillators featured with blight and fast light and self-absorption, such as ZnO, PbI_2 , GaAs/ InAs etc., may function as quantum dots in composite scintillators with tunable emission and no afterglow for fast scintillator screens.
- ❑ Require matching fast photodetector, e.g. SiPM or LAPPD, and fast electronics.
- ❑ Type I : BaF_2 based screens: 3 years.
- ❑ Type II: Wide-band semiconductor QD based scintillator screens: 5 years.





- ❑ Caltech HEP crystal lab worked on crystal development for HEP calorimeters, such as L3 BGO, BaBar CsI(Tl) and CMS PWO, and is working on Mu2e CsI as well as LYSO and BaF_2 crystals for future HEP experiments at the energy and intensity frontiers.
- ❑ Existing instruments and facilities for crystal characterization and radiation damage tests:
  - Edinburgh Instrument FLS spectrometer.
  - PerkinElmer L950 UV/Vis/NIR spectrophotometer.
  - Hitachi U3210 and F4500 spectrophotometers.
  - 20 GS digital scope and LeCroy QVTs.
  - Nano second OPO laser, excitation sources.
  - Co-60, Cs137 and Cf-252 source facilities.

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