



Recent Progress on Fast Inorganic Scintillators for Future HEP Experiments Ren-Yuan Zhu California Institute of Technology December 7, 2017

Technical Seminar Presented at Fermi National Laboratory



Fast & Radiation Hard Scintillators



- Supported by the DOE ADR program we are developing fast and radiation hard scintillators to face the challenge for future HEP experiments at the energy and intensity frontiers.
- LYSO:Ce, BaF₂ and LuAG:Ce will survive the radiation environment expected at HL-LHC with 3000 fb⁻¹. LYSO is proposed for a MIP timing detector for CMS upgrade:
 - Absorbed dose: up to 100 Mrad,
 - Charged hadron fluence: up to 6×10¹⁴ p/cm²,
 - Fast neutron fluence: up to 3×10¹⁵ n/cm².
- Ultra-fast scintillators with excellent radiation hardness is also needed to face the challenge of unprecedented event rate expected at future HEP experiments at the intensity frontier, such as Mu2e-II, and the GHz X-ray imaging for the proposed Marie project at Los Alamos. Y:BaF₂ with a sub-ns FWHM pulse width and a suppressed slow scintillation component is a leading candidate for both applications.

Bright & Fast Scintillators: LYSO & BaF₂



Crystal	Nal(TI)	CsI(TI)	Csl	BaF ₂	BGO	LYSO(Ce)	PWO	PbF ₂
Density (g/cm ³)	3.67	4.51	4.51	4.89	7.13	7.40	8.3	7.77
Melting Point (°C)	651	621	621	1280	1050	2050	1123	824
Radiation Length (cm)	2.59	1.86	1.86	2.03	1.12	1.14	0.89	0.93
Molière Radius (cm)	4.13	3.57	3.57	3.10	2.23	2.07	2.00	2.21
Interaction Length (cm)	42.9	39.3	39.3	30.7	22.8	20.9	20.7	21.0
Refractive Index ^a	1.85	1.79	1.95	1.50	2.15	1.82	2.20	1.82
Hygroscopicity	Yes	Slight	Slight	No	No	No	No	No
Luminescence ^b (nm) (at peak)	410	550	310	300 220	480	402	425 420	?
Decay Time ^b (ns)	245	1220	26	650 0.9	300	40	30 10	?
Light Yield ^{b,c} (%)	100	165	4.7	36 4.1	21	85	0.3 0.1	?
d(LY)/dT ^ь (%/ ºC)	-0.2	0.4	-1.4	-1.9 0.1	-0.9	-0.2	-2.5	?
Experiment	Crystal Ball	BaBar BELLE BES-III	KTeV S.BELLE Mu2e-I	(GEM) TAPS Mu2e-II	L3 BELLE HHCAL?	COMET & CMS (Mu2e & SperB)	CMS ALICE PANDA	A4 g-2 HHCAL

a. at peak of emission; b. up/low row: slow/fast component; c. QE of readout device taken out.

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Light Output & Decay Kinetics



Measured with Philips XP2254B PMT (multi-alkali cathode) p.e./MeV: LSO/LYSO is 6 & 230 times of BGO & PWO respectively



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Fast Signals with 1.5 X₀ Samples



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



The 3 ns width of BaF₂ pulse may be further reduced by faster photodetector LYSO, LaBr₃ & CeBr₃ have tail, which would cause pile-up for GHz readout

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Fast Inorganic Scintillators for HEP



	LYSO:Ce	LSO:Ce, Ca ^[1]	LuAG:Ce	LuAG:Pr ^[3]	GGAG:Ce ^[4,5]	Csl	BaF ₂ ^[6]	BaF ₂ :Y	CeBr ₃	LaBr ₃ :Ce ^[7]
Density (g/cm ³)	7.4	7.4	6.76	6.76	6.5	4.51	4.89	4.89	5.23	5.29
Melting points (°C)	2050	2050	2060	2060	1850 ^d	621	1280	1280	722	783
X _o (cm)	1.14	1.14	1.45	1.45	1.63	1.86	2.03	2.03	1.96	1.88
R _M (cm)	2.07	2.07	2.15	2.15	2.20	3.57	3.1	3.1	2.97	2.85
λ _ι (cm)	20.9	20.9	20.6	20.6	21.5	39.3	30.7	30.7	31.5	30.4
Z _{eff}	64.8	64.8	60.3	60.3	51.8	54.0	51.6	51.6	45.6	45.6
dE/dX (MeV/cm)	9.55	9.55	9.22	9.22	8.96	5.56	6.52	6.52	6.65	6.90
λ _{peak} ^a (nm)	420	420	520	310	540	310	300 220	300 220	371	360
PL Emission Peak (nm)	402	402	500	308	540	310	300 220	300 220	350	360
PL Excitation Peak (nm)	358	358	450	275	445	256	<200	<200	330	295
Absorption Edge (nm)	170	170	160	160	190	200	140	140	n.r.	220
Refractive Index ^b	1.82	1.82	1.84	1.84	1.92	1.95	1.50	1.50	1.9	1.9
Normalized Light Yield ^{a,c}	100	116 ^e	35 ^f 48 ^f	44 41	40 75	4.2 1.3	42 5.0	1.7 5.0	99	153
Total Light yield (ph/MeV)	30,000	34,800°	25,000 ^f	25,800	34,700	1,700	13,000	2,100	30,000	46,000
Decay time ^a (ns)	40	31 ^e	981 ^f 64 ^f	1208 26	319 101	30 6	600 <mark>0.6</mark>	600 <mark>0.6</mark>	17	20
Light Yield in 1 st ns (photons/MeV)	740	950	240	520	260	100	1200	1200	1,700	2,200
Issues					neutron x-section	Slightly hygroscop ic	Slow compon ent	DUV PD	hygr	oscopic

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Fast Inorganic Scintillators (II)



a. Top line: slow component, bottom	[1] Spurrier, et al., <i>IEEE T. Nucl. Sci.</i> 2008,55 (3):				
line: fast component;	1178-1182				
b. At the wavelength of the emission	[2] Liu, et al., Adv. Opt. Mater. 2016, 4(5): 731–739				
mavimum	[3] Hu, et al., <i>Phys. Rev. Applied</i> 2016, 6: 064026				
	[4] Lucchini, et al., <i>NIM A</i> 2016, 816: 176-183				
c. Excited by Gamma rays;	[5] Meng, et al., <i>Mat. Sci. Eng. B-Solid</i> 2015, 193:				
d. For Gd ₃ Ga ₃ Al ₂ O ₁₂ :Ce	20-26				
e. For 0.4 at% Ca co-doping	[6] Diehl, et al., <i>J. Phys. Conf. Ser</i> 2015, 587:				
f. Ceramic with 0.3 Mg at% co-doping	012044				
	[7] Pustovarov, et al., Tech. Phys. Lett. 2012, 784-				
	788				



LuAG:Ce Ceramic Samples



Presented in NSS2017, will be published in IEEE TNS NS



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Excellent Radiation Hardness



No damage observed in both transmittance and light output after 220 Mrad ionization dose and 3×10^{14} p/cm² of 800 MeV



Very promising for a scintillating ceramics based calorimeter Presented in NSS2017 will be published in IEEE TNS NS



- **Crystal lateral dimension:** $\pm 100 \mu$, length: $\pm 100 \mu$.
- ❑ Scintillation properties at seven points along the crystal wrapped by two layers of Tyvek paper of 150 µm for alternative end coupled to a bi-alkali PMT with an air gap. Light output and FWHM resolution are the average of seven points with 200 ns integration time. The light response uniformity is the rms of seven points. F/T is measured at the point of 2.5 cm to the PMT.
 - Light output (LO): > 100 p.e./MeV with 200 ns gate, will be compared to reference for cross-calibration;
 - □ FWHM Energy resolution: < 45% for Na-22 peak;
 - Light response uniformity (LRU, rms of seven points): < 5%;</p>
 - □ Fast (200 ns)/Total (3000 ns) Ratio: > 75%.
- □ Radiation related spec::
 - Normalized LO after 10/100 krad: > 85/60%;
 - □ Radiation Induced noise @ 1.8 rad/h: < 0.6 MeV.



Mu2e Preproduction Csl



A total of 72 crystals from Amcrys, Saint-Gobain and SICCAS has been measured at Caltech and LNF

Amerys C0013	S-G C0045	SIC C0037
Amerys C0015	S-G C0046	SIC C0038
Amerys C0016	S-G C0048	SIC C0039
Amerys C0019	S-G C0049	SIC C0040
Amerys C0023	S-G C0051	SIC C0041
Amcrys C0025	S-G C0057	SIC C0042
Amerys C0026	S-G C0058	SIC C0043
Amerys C0027	S-G C0060	SIC C0068
Amerys C0030	S-G C0062	SIC C0070
Amerys C0032	S-G C0063	SIC C0071
Amerys C0034	S-G C0065	SIC C0072
Amerys C0036	S-G C0066	SIC C0073

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Quality of Pre-Production Csl



Presented in SCINT2017, will be published in IEEE TNS NS



Most preproduction crystals satisfy specifications, except a few crystals from SICCAS fail the LRU spec and about half Amcrys crystals fail the F/T ratio and RIN

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Fast and Slow Light from BaF₂

A radiation level exceeding 100 krad is expected at the proposed Mu2e-II, so BaF₂ is being considered.

The amount of light in the fast component of BaF₂ at
220 nm with sub-ns decay time is similar to CsI.

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





Slow Suppression: Doping & Readout



Slow component may be suppressed by RE doping: Y, La and Ce

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.



Solar-blind cathode (Cs-Te) + La doping achieved F/S = 5/1



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Eight BaF₂ Plates from BGRI



ID	Dimension (mm ³)	Polishing					
BGRI Y-doped BaF ₂ -1708-1,4	10x10x2	Double faces					
BGRI Undoped BaF ₂ -1708-5,8	Double faces						
All samples received in August 31, 2017							

Experiments

Properties measured at room temperature : PHS, LO & Decay kinetics







Good transparency for both fast and slow scintillation





Light Output: Y:BaF₂ and BaF₂



F/S ratio increased from 0.21 to 6 .2, presented in NSS2017



Being irradiation up to 200 Mrad and 2x10¹⁵ n/cm² at the East Port of LANSCE



Pulse Shape: BaF₂ Cylinders



BGRI BaF₂ cylinders of Φ10×10 cm³ shows γ-ray response: 0.26/0.55/0.94 ns of rising/decay/FWHM width





Tail Reduced in BGRI BaF₂:Y



Slow component tail observed in 2 µs in BaF2, not BaF2:Y



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Summary of BaF₂ Cylinders



Consistent pulse shape observed between PbF_2 and BaF_2 , indicating that the decay time of the fast component in BaF_2 less than 0.6 ns, faster than literature

Samples	Dimensions	Excitation	Rise time (ns)	Decay time (ns)	FWHM (ns)
MCP- PMT240*	Φ40 mm	Laser pulse	0.185	N/A	1.36
PbF	50×50×50 mm ³	Cosmic-ray	0.18±0.05	0.61±0.05	0.88±0.07
SIC-U	Φ10×10 mm ³	Cosmic-ray	0.26±0.05	0.52±0.05	0.92±0.07
SIC-Y	Φ10×10 mm ³	Cosmic-ray	0.26±0.05	0.57±0.05	0.98±0.07
BGRI-U	Φ10×10 mm ³	Na-22 (511KeV)	0.22±0.05	0.59±0.05	0.92±0.07
BGRI-Y	Φ10×10 mm ³	Na-22 (511KeV)	0.29±0.05	0.50±0.05	0.96±0.07

*From test report of the Photek PMT240 MCPT.

BGRI/Incrom/SIC BaF₂ Samples







ID	Vendor	Dimension (mm ³)	Polishing
SIC 1-20	SICCAS	30x30x250	Six faces
BGRI-2015 D, E, 511	BGRI	30x30x200	Six faces
Russo 2, 3	Incrom	30x30x200	Six faces



BaF₂: Normalized EWLT and LO



Consistent damage in crystals from three vendors



Remaining light output after 120 Mrad: 40%/45% for the fast/slow component



RIAC & LO Vs. Proton Fluence

Excellent radiation hardness of LYSO and BaF_2 up to 10^{15} p/cm²



Presented in SCINT 2017, will be published in IEEE TNS NS

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Neutron Irradiation Test at LANL



Samples are placed at the Target-4

Los Alamos Neutron Science Center (LANSCE)

East Port, about 1.2 m away from Bldg. the neutron production target. 823 Line D Proton Beam Control Room/ Data Bldg. 541 Acquisition East Port Bldg. 7 Target 2 Bldg. Blue Room 1138 2FP120L 2FP60R Sample Holder Target 2 Target-4 4FP60R oture Bldg. 382 Bldg. 370 4FP30R Vacuum Neutron Production Target Θ ICE House 4FP15B Bldg. 1265 Bldg. 371 50 cm Bldg. 372 4FP30L Concrete & Steel Shielding Bldg. 34 Bldg 616 4FP15L

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Neutrons/Photons/Protons Fluxes



Neutrons/Photons/Protons fluxes are calculated by using MCNPX (Monte Carlo N-Particle eXtended). Plotted spectra are tallied in the largest sample volume (averaging)







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

Organizers: Peter Denes, Sol Gruner, Michael Stevens & Zhehui (Jeff) Wang¹ (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 Los Alamos National Laboratory Report LA-UR-17-22085 (2017). ultrafast hard X-ray imaging the too barren be and applications in the next 5 to 10 years, see Table 1, and to establish the foundations for near-term R&D collaborations.

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

Table I. High-energy photon imagers for MaRIE XFEL

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



Why Crystal Scintillator?



LANL Report LA-UR-17-22085 (2017)

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

Ultra-fast crystals, photodetectors and readout.

X-ray photons

Fast Electronics

Photo

Fast Scintillator

Screen

Detectors



Pixelized Crystal Detectors



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





Laser slicing, micropore or not pixelized provide better coverage



Candidate Scintillators for Marie

Proc. of SPIE 10392 (2017)



					`					
	LYSO (:Ce)	YSO:Ce	ZnO:Ga	BaF ₂	BaF ₂ :Y	YAP:Ce	YAP:Yb	YAG:Yb	LuAG:Ce	LaBr ₃ (:Ce)
Density (g/cm ³)	7.4	4.44	5.67	4.89	4.89	5.35	5.35	4.56	6.76	5.29
Melting points (°C)	2050	2070	1975	1280	1280	1870	1870	1940	2060	783
X ₀ (cm)	1.14	3.10	2.51	2.03	2.03	2.77	2.77	3.53	1.45	1.88
R _M (cm)	2.07	2.93	2.28	3.1	3.1	2.4	2.4	2.76	2.15	2.85
λ _ι (cm)	20.9	27.8	22.2	30.7	30.7	22.4	22.4	25.2	20.6	30.4
Z _{eff}	64.8	33.3	27.7	51.6	51.6	31.9	31.9	30	60.3	45.6
dE/dX (MeV/cm)	9.55	6.57	8.42	6.52	6.52	8.05	8.05	7.01	9.22	6.90
λ _{peak} ^a (nm)	420	420	389	300 220	300 220	370	350	350	520	360
Refractive Index ^b	1.82	1.78	2.1	1.50	1.50	1.96	1.96	1.87	1.84	1.9
Normalized Light Yield ^{a,c}	100	80	6.6 ^e	42 4.8	1.7 4.8	9 32	0.19 ^e	0.36 ^e	35 ^f 48 ^f	153
Total Light yield (ph/MeV)	30,00 0	24,000	2,000 ^e	13,000	2,000	12,000	57°	110 e	25,000 ^f	46,000
Decay time ^a (ns)	40	75	<1	600 <mark>0.6</mark>	600 <mark>0.6</mark>	191 25	1.5	4	981 ^f 64 ^f	20
Light Yield in 1 st ns (photons/MeV)	740	318	610 ^e	1200	1200	391	28 ^e	24 ^e	240	2,200
40 keV Att. Length (1/e, mm)	0.185	0.334	0.407	0.106	0.106	0.314	0.314	0.439	0.251	0.131
[1] Spurrier, et al., <i>IEEE T. Nucl. Sci.</i> 2008,55 (3): 1178-1192.						c. Excited by Gamma rays;				
b. At the wavele	ngth of th	ne emission ma	iximum;	Jonent,		e. Excited by Alpha particles.				

f. Ceramic with 0.3 Mg at% co-doping

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LYSO and ZnO:Ga Samples





Crystal	Vendor	ID	Dimension (mm ³)
LYSO:Ce	SIC	150210-1	19x19×2
YSO:Ce	SIC	51	25×25×5
ZnO:Ga	FJIRSM	2014-1	33×30×2
ZnO:Ga	FJIRSM	2014-2	22×22×0.3

Experiments

 Properties measured at room temperature : PL & Decay, Transmittance, PHS, LO & Decay kinetics



FJIRSM 2mm ZnO:Ga-2014-1



Very short decay time

× Low EWLT and LO due to severe self absorption



ID	Dimension	EWLT (%)	ER (%)	50 ns LO (p.e./MeV)	Primary Decay Time (ns)
FJIRSM ZnO:Ga-2014-1	33×30×2	7.0	37.8	76 (α)	2.7



FJIRSM 0.3 mm ZnO:Ga-2014-2



× Reduced self absorption due to 0.3 mm thickness

× May pursue QD, NP or thin film based solution



ID	Dimension	EWLT (%)	ER (%)	50 ns LO (p.e./MeV)	Primary Decay Time (ns)
FJIRSM ZnO:Ga-2014-2	22×22×0.3	10.8	18.2	296 (α)	3.5



ZnO:Ga Polystyrene Composite



P. Lecoq, Talk in the Picosecond workshop, Kansas City, 15-18 September, 2016

- Highly luminescent ZnO:Ga nano crystals 80-100nm
 - Prepared by a photochemical method
 - Embedded in a polystyrene sheet 10%weigth





BaF₂ and Other Samples





Crystal	Vendor	Vendor ID	
BaF ₂	SIC	1	50×50×5
BaF ₂ :Y	BGRI	1708	10×10×2
YAP:Ce	Dongjun	2102	Ф50×2
YAP:Yb	Dongjun	2-2	Ф40×2
YAG:Yb	Dongjun	4	10×10×5
LuAG:Ce	SIC	S2	25×25×0.4

Experiments

 Properties measured at room temperature : PL & Decay, Transmittance, PHS, LO & Decay kinetics



SIC BaF₂-1



The highest LY in 1st ns among all non-hygroscopic scintillators

× ~600 ns slow component may be suppressed by Y doping



ID	Dimension	EWLT (%)	ER (%)	50 ns LO (p.e./MeV)	Primary Decay Time (ns)
SIC BaF ₂ -1	50×50×5	85.1	54.9	209	0.6



Use Thin Layer Scintillators



Proc. of SPIE 0504 (2015)

A multilayer high QE photocathode coated thin fast scintillators concept was proposed for GHz hard X-ray imaging:

- Spatial resolution determined layer thickness,
- Overall efficiency defined layer number,
- Maximized conversion of scintillation photon to p.e.,
- Magnetic field extraction of p.e. and image preserving,
- Off-beam p.e. multiplication,
- On-board charge storages.



Figure 6. A multi-layer detector architecture for efficient and fast imaging of diffracted X rays. A guide magnetic field perpendicular to the X-ray direction guide the photoelectrons to amplification and storage. The magnetic field also preserves the image contrast due to X-ray absorption at the scintillator location.



Ag/Au-ZnO Core-Shell Nano Particles



Nature Scientific **Reports** | 5:14004 | DOI: 10.1038/srep14004



Figure 2. SEM images for ZnO samples without nanoparticles (**a**), with 2 mL_AgNP (**b**), 8 mL_AgNP (**c**), and 8 mL_AuNP (**d**) illustrating the change in shape of the particles. The particles in the lower two micrographs are referred to as "star" or "thistle" shaped in the text.

Presentation by Ren-Yuan Zhu, Caltech, in Fermilab

Enhanced UV Emission in Ag/Au-ZnO

Nature Scientific Reports | 5:14004 | DOI: 10.1038/srep14004

- Enhancement of ZnO near-bandedge (UV) emission centered at 385 nm was reported in PL and RL of Ag/Au-ZnO core shell nanoparticles.
- The enhanced luminescence and the decreased free exciton lifetime suggest a plasmon-coupled-emission mechanism.
- This suggests that plasmon-coupled luminescence can be employed for the development of improved scintillators.







Summary: HEP Experiments



- LYSO, BaF₂ crystals and LuAG ceramics show an excellent radiation hardness beyond 100 Mrad, 1 x 10¹⁵ p/cm² and 2 x 10¹⁵ n/cm². They promise a very fast and robust detector in a severe radiation environment, such as the HL-LHC.
- □ Commercially available undoped BaF₂ crystals provide sufficient fast light with sub-ns FWHM pulse width. Yttrium doping in BaF₂ crystals increases its F/S ratio from 1/5 to 6/1 while maintaining the intensity of its sub-ns fast component. The slow contamination at this level is already less than commercially available undoped CsI, so is promising for Mu2e-II and GHz X-ray imaging.
- Results of the experiments 6991 and 7332 at LANL show fast neutrons up to 2 x 10¹⁵ n/cm² do not damage LYSO, BaF₂ and PWO crystals, confirming early observation at Saclay reactor.
- Our plan is to investigate LYSO:Ce,Ca crystals, LuAG:Ce and LuAG:Pr ceramics and try to develop Y:BaF₂ crystals for future HEP experiments. Will also pay an attention to photodetector with DUV response: LAPPD, Si or diamond based solid state detectors.



Hamamatsu S13371-6050CQ-02



SiPM with VUV response is available: QE = 22% at 220 nm





Diamond Photodetector



In addition to SiPM with VUV response

E. Monroy, F. Omnes and F. Calle,"Wide-bandgap semiconductor ultraviolet photodetectors, IOPscience 2003 Semicond. Sci. Technol. 18 R33





Figure 6. Quantum efficiency of diamond photoconductors at different temperatures and Arrhenius plot of the peak value (inset). (From [Sal00].)

Fig.4. External quantum efficiency extended to visible and near infrared wavelength regions. The

E. Pace and A. De Sio, "Innovative diamond photo-detectors for UV astrophysics", Mem. S.A.It. Suppl. Vol. 14, 84 (2010)



Summary: GHz Imaging



- GHz hard X-ray imaging for the proposed Marie project presents an unprecedented challenge to the speed and radiation hardness of the inorganic scintillators.
- BaF₂ crystals provide sufficient fast light with a sub-ns FWHM pulse width and an excellent radiation hardness beyond 100 Mrad and 1 x 10¹⁵ h/cm². With its slow component effectively suppressed by yttrium doping Y:BaF₂ promises a fast and robust front imager.
- Bulk ZnO:Ga crystals suffer from serious self-absorption. Enhanced UV emission observed in Ag/Au ZnO core-shell nano particles hints a thin film based approach.
- Our plan is to investigate along both lines: Y:BaF₂ crystals, and ZnO QD/NP based thin film for the Marie project with a close collaboration between the NP, HEP and material science community.