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# Applications of Very Fast Inorganic Crystal Scintillators for Future HEP Experiments

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# 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 crystals will survive the radiation environment expected at HL-LHC with  $3000 \text{ fb}^{-1}$ , so are proposed for a precision timing layer for CMS upgrade:**
  - Absorbed dose: up to 100 Mrad,
  - Charged hadron fluence: up to  $6 \times 10^{14} \text{ p/cm}^2$ ,
  - Fast neutron fluence: up to  $3 \times 10^{15} \text{ n/cm}^2$ .
- **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. Y:BaF<sub>2</sub> with sub-ns decay time and suppressed slow scintillation component is a leading candidate for both applications.**



# Bright & Fast Scintillators: LYSO & BaF<sub>2</sub>



Crystal	NaI(Tl)	CsI(Tl)	CsI	BaF <sub>2</sub>	BGO	LYSO(Ce)	PWO	PbF <sub>2</sub>
Density (g/cm <sup>3</sup> )	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 <sup>a</sup>	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 <sup>b</sup> (nm) (at peak)	410	550	310	300 220	480	402	425 420	?
Decay Time <sup>b</sup> (ns)	245	1220	26	650 0.9	300	40	30 10	?
Light Yield <sup>b,c</sup> (%)	100	165	4.7	36 4.1	21	85	0.3 0.1	?
d(LY)/dT <sup>b</sup> (%/°C)	-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.

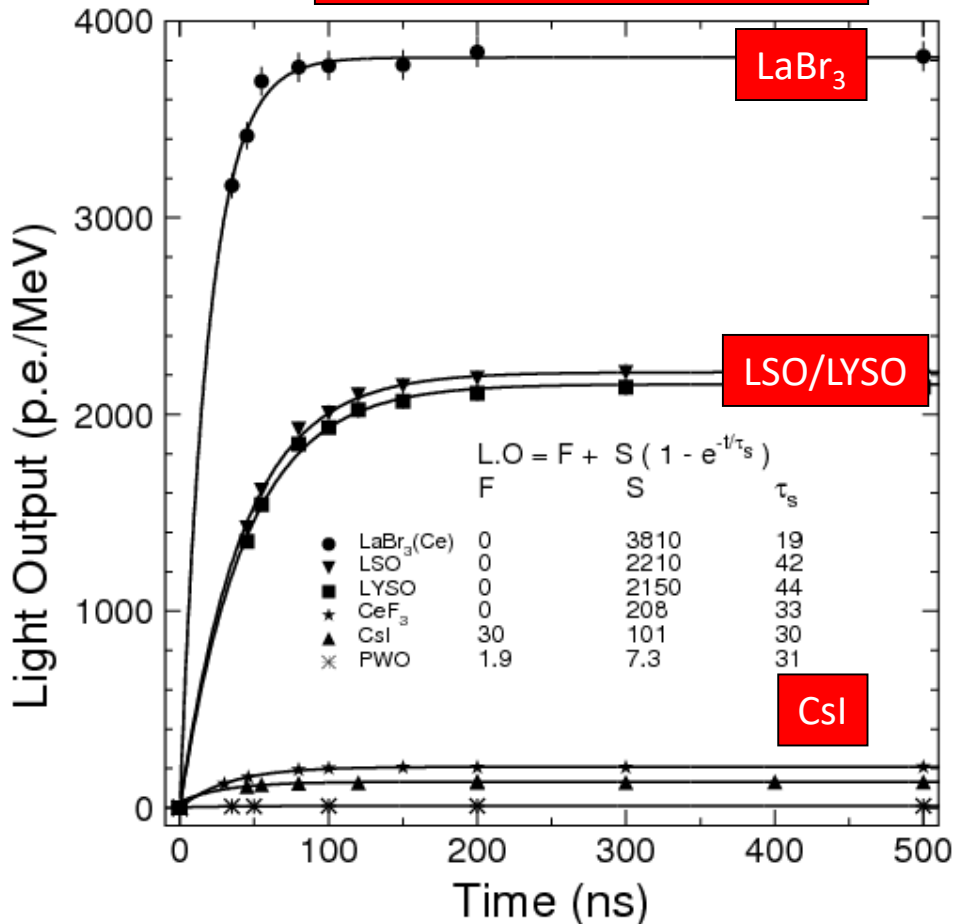


# Light Output & Decay Kinetics

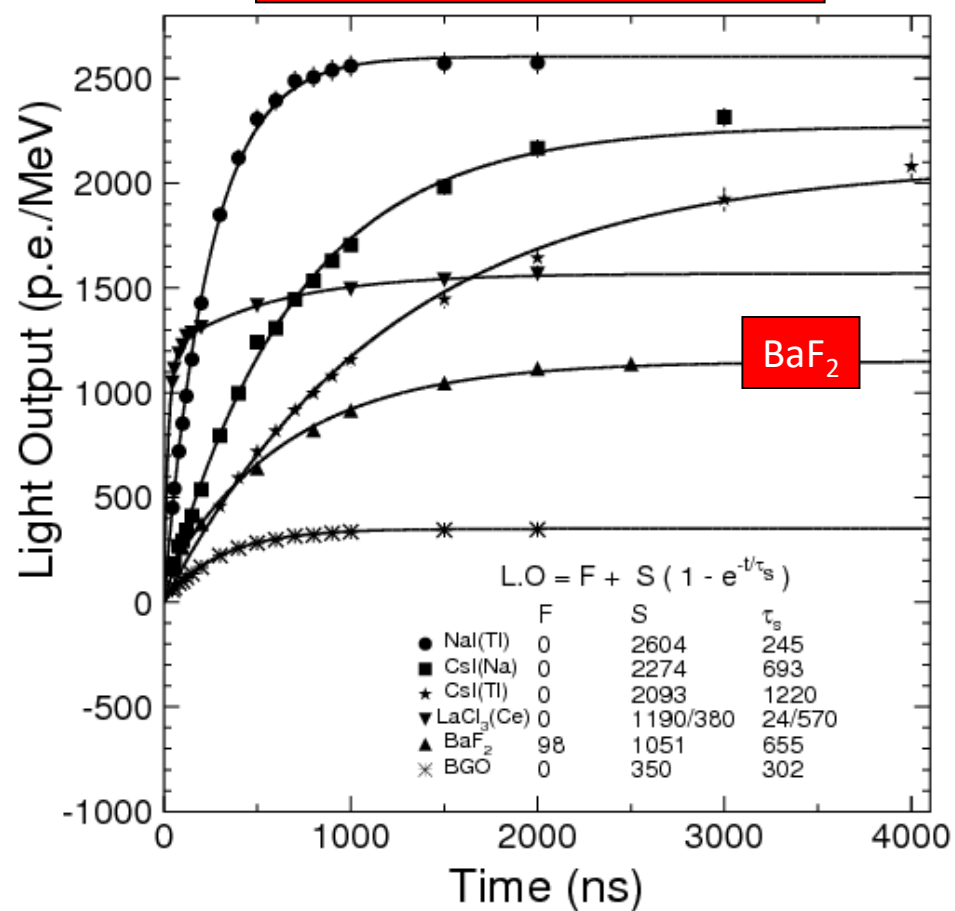


Measured with Philips XP2254B PMT (multi-alkali cathode)  
 p.e./MeV: LSO/LYSO is 6 & 230 times of BGO & PWO respectively

Fast Crystal Scintillators



Slow Crystal Scintillators

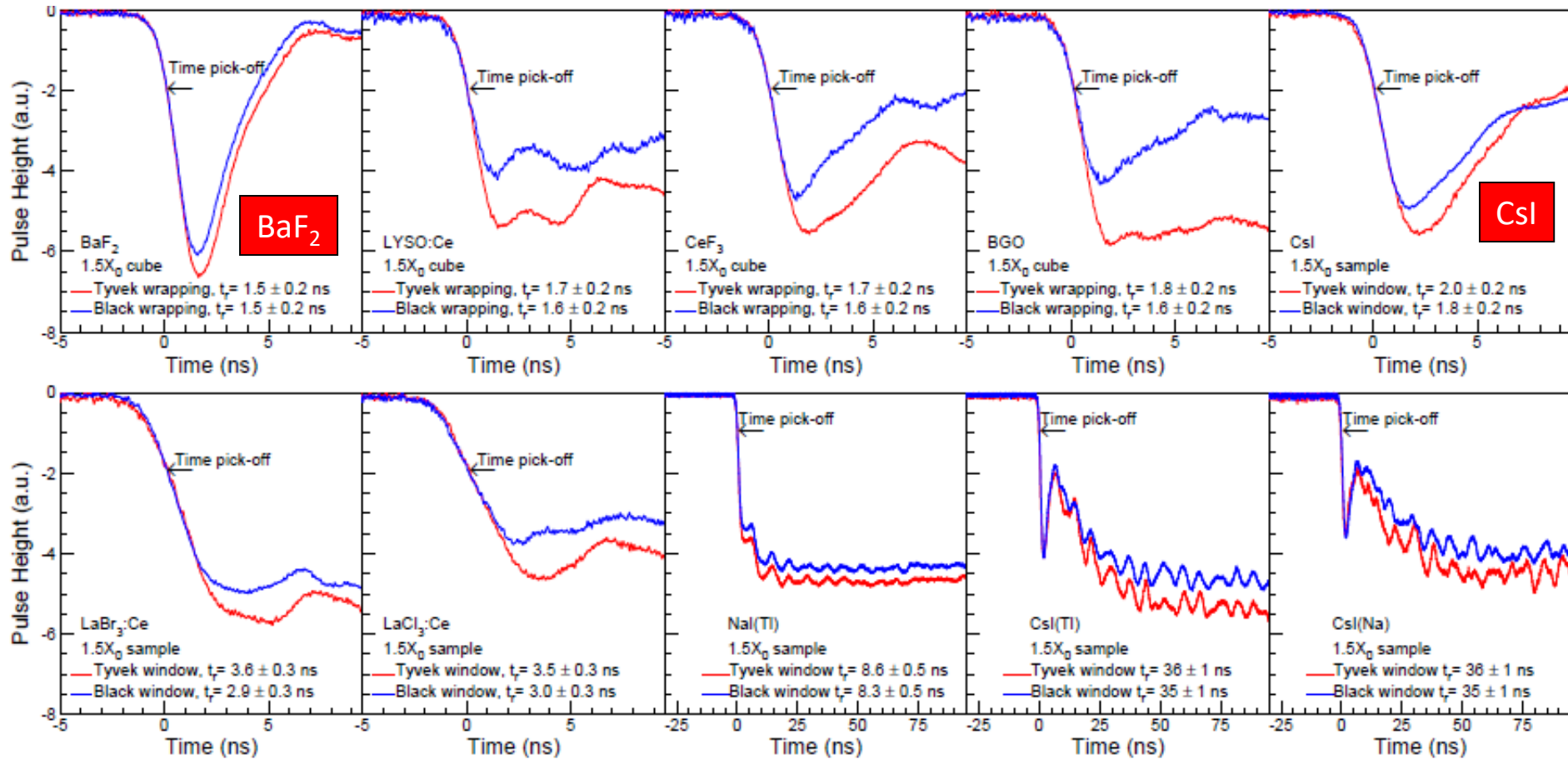




# Fast Signals: $1.5 X_0$ Samples



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



The narrow width of BaF<sub>2</sub> pulse may be reduced by a faster photodetector  
LYSO, LaBr<sub>3</sub> & CeBr<sub>3</sub> have tail, which would cause pile-up for GHz readout



# Figure of Merit for Timing



FoM is calculated as the LY in 1<sup>st</sup> ns obtained by using light output and decay time data measured for 1.5 X<sub>0</sub> crystal samples.

Crystal Scintillators	Relative LY (%)	A <sub>1</sub> (%)	τ <sub>1</sub> (ns)	A <sub>2</sub> (%)	τ <sub>2</sub> (ns)	Total LO (p.e./MeV, XP2254B)	LO in 1ns (p.e./MeV, XP2254B)	LO in 0.1ns (p.e./MeV, XP2254B)	LY in 0.1ns (photons/MeV)
BaF <sub>2</sub>	40.1	91	650	9	0.9	1149	71.0	11.0	136.6
LSO:Ca,Ce	94	100	30			2400	78.7	8.0	110.9
LSO/LYSO:Ce	85	100	40			2180	53.8	5.4	75.3
CeF <sub>3</sub>	7.3	100	30			208	6.8	0.7	8.6
BGO	21	100	300			350	1.2	0.1	2.5
PWO	0.377	80	30	20	10	9.2	0.42	0.04	0.4
LaBr <sub>3</sub> :Ce	130	100	20			3810	185.8	19.0	229.9
LaCl <sub>3</sub> :Ce	55	24	570	76	24	1570	49.36	5.03	62.5
NaI:Tl	100	100	245			2604	10.6	1.1	14.5
CsI	4.7	77	30	23	6	131	7.9	0.8	10.6
CsI:Tl	165	100	1220			2093	1.7	0.2	4.8
CsI:Na	88	100	690			2274	3.3	0.3	4.5

The best crystal scintillator for ultra-fast rate is BaF<sub>2</sub> and LSO(Ce/Ca) and LYSO(Ce). LaBr<sub>3</sub> is a material with high potential.



# Fast Inorganic Scintillators



	LYSO:Ce	LSO:Ce, Ca <sup>[1]</sup>	LuAG:Ce <sub>[2]</sub>	LuAG:Pr <sup>[3]</sup>	GGAG:Ce <sup>[4,5]</sup>	CsI	BaF <sub>2</sub> <sup>[6]</sup>	BaF <sub>2</sub> :Y	CeBr <sub>3</sub>	LaBr <sub>3</sub> :Ce <sup>[7]</sup>
Density (g/cm <sup>3</sup> )	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 <sup>d</sup>	621	1280	1280	722	783
X <sub>0</sub> (cm)	1.14	1.14	1.45	1.45	1.63	1.86	2.03	2.03	1.96	1.88
R <sub>M</sub> (cm)	2.07	2.07	2.15	2.15	2.20	3.57	3.1	3.1	2.97	2.85
λ <sub>i</sub> (cm)	20.9	20.9	20.6	20.6	21.5	39.3	30.7	30.7	31.5	30.4
Z <sub>eff</sub>	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 <sub>peak</sub> <sup>a</sup> (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 <sup>b</sup>	1.82	1.82	1.84	1.84	1.92	1.95	1.50	1.50	1.9	1.9
Normalized Light Yield <sup>a,c</sup>	100	116 <sup>e</sup>	35 <sup>f</sup> 48 <sup>f</sup>	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 <sup>e</sup>	25,000 <sup>f</sup>	25,800	34,700	1,700	13,000	2,100	30,000	46,000
Decay time <sup>a</sup> (ns)	40	31 <sup>e</sup>	981 <sup>f</sup> 64 <sup>f</sup>	1208 26	319 101	30 6	600 <b>0.6</b>	600 <b>0.6</b>	17	20
Light Yield in 1 <sup>st</sup> ns (photons/MeV)	740	950	240	520	260	100	1200	1200	1,700	2,200
Issues					neutron x-section	Slightly hygroscopic	Slow component	DUV PD	hygroscopic	



# Fast Inorganic Scintillators (II)



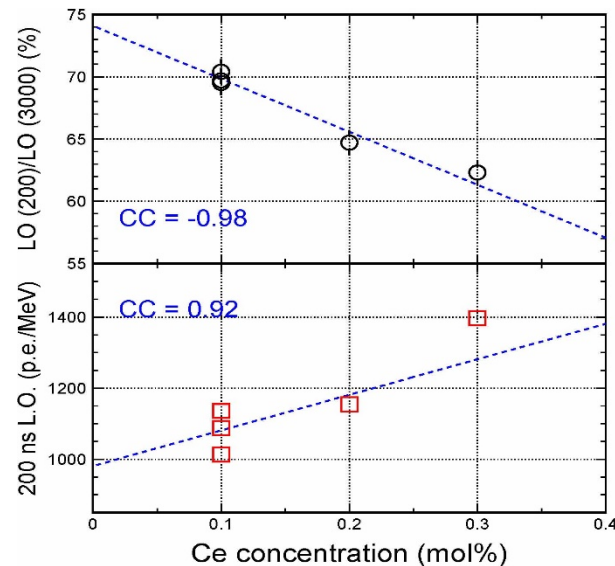
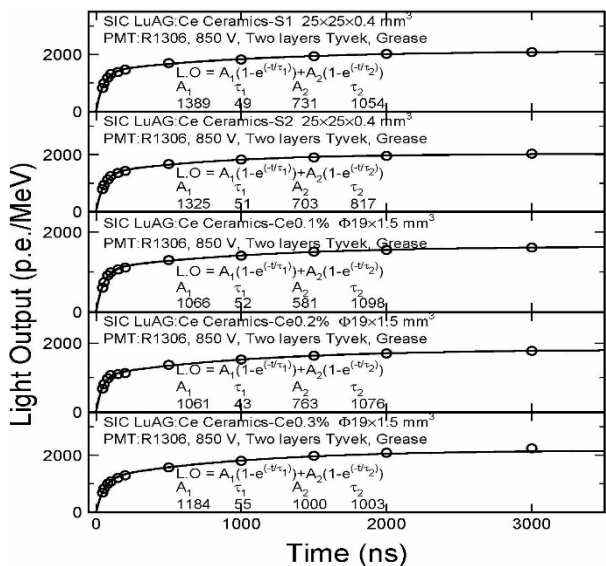
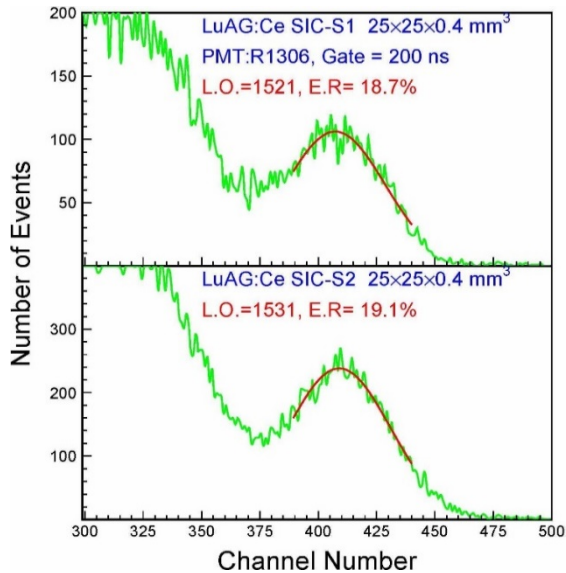
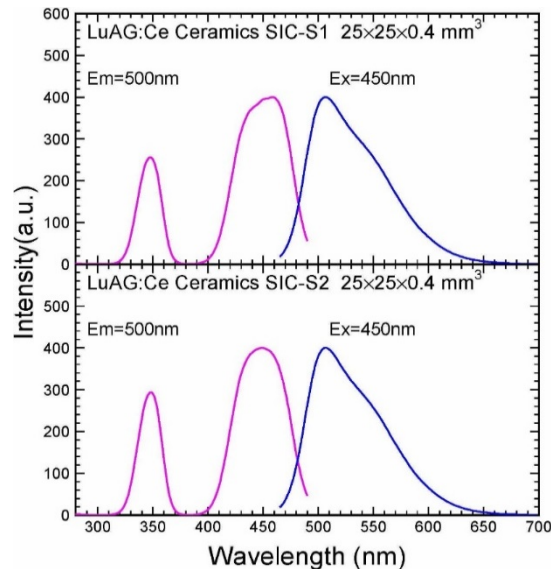
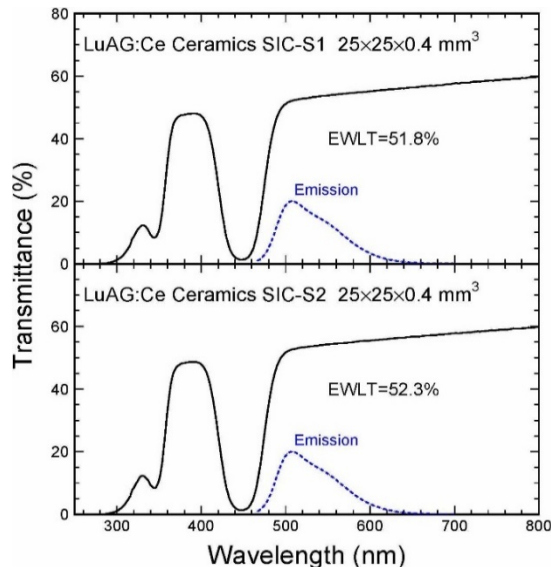
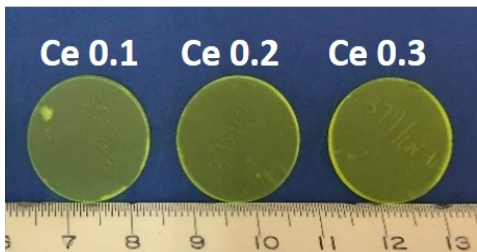
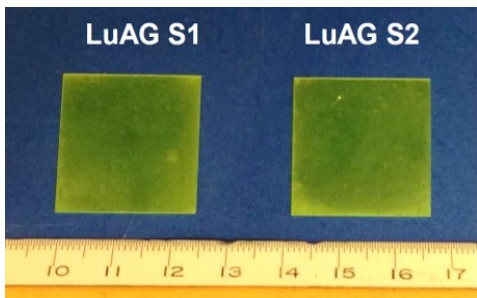
- a. Top line: slow component, bottom line: fast component;**
- b. At the wavelength of the emission maximum;**
- c. Excited by Gamma rays;**
- d. For  $\text{Gd}_3\text{Ga}_3\text{Al}_2\text{O}_{12}:\text{Ce}$**
- e. For 0.4 at% Ca co-doping**
- f. Ceramic with 0.3 Mg at% co-doping**

- [1] Spurrier, et al., *IEEE T. Nucl. Sci.* 2008,55 (3): 1178-1182
- [2] Liu, et al., *Adv. Opt. Mater.* 2016, 4(5): 731–739
- [3] Hu, et al., *Phys. Rev. Applied* 2016, 6: 064026
- [4] Lucchini, et al., *NIM A* 2016, 816: 176-183
- [5] Meng, et al., *Mat. Sci. Eng. B-Solid* 2015, 193: 20-26
- [6] Diehl, et al., *J. Phys. Conf. Ser* 2015, 587: 012044
- [7] Pustovarov, et al., *Tech. Phys. Lett.* 2012, 784-788





# LuAG:Ce Ceramic Samples

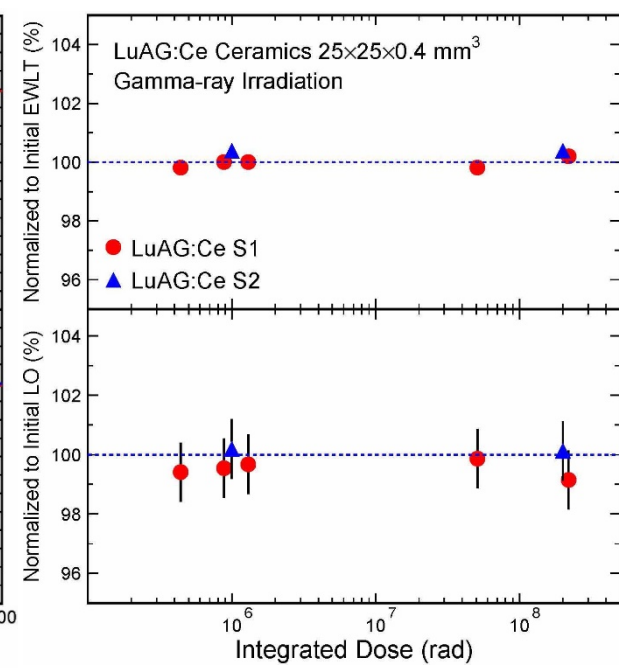
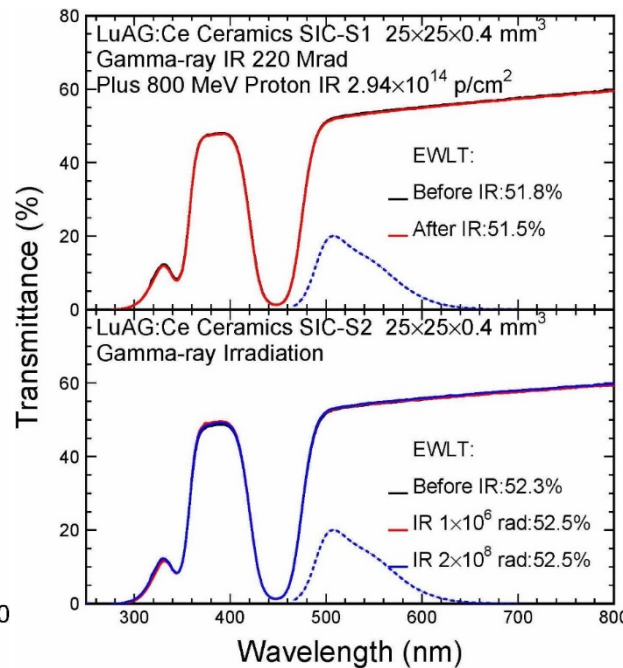
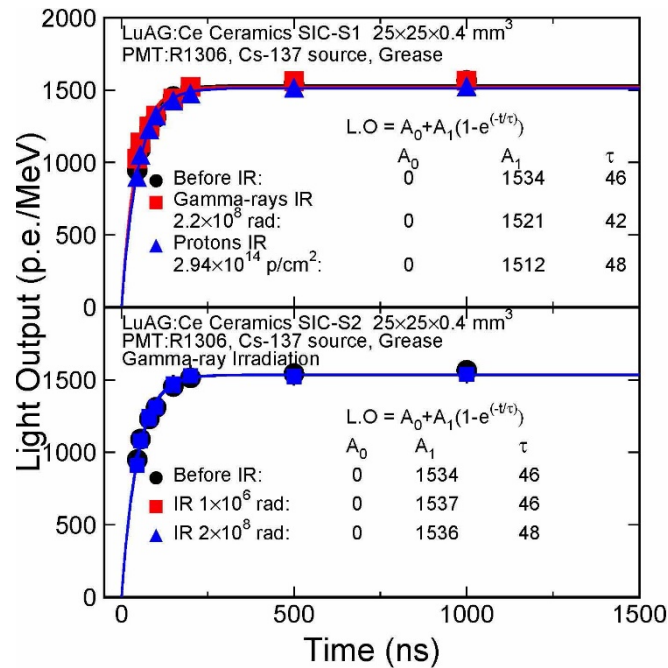




# Excellent Radiation Hardness



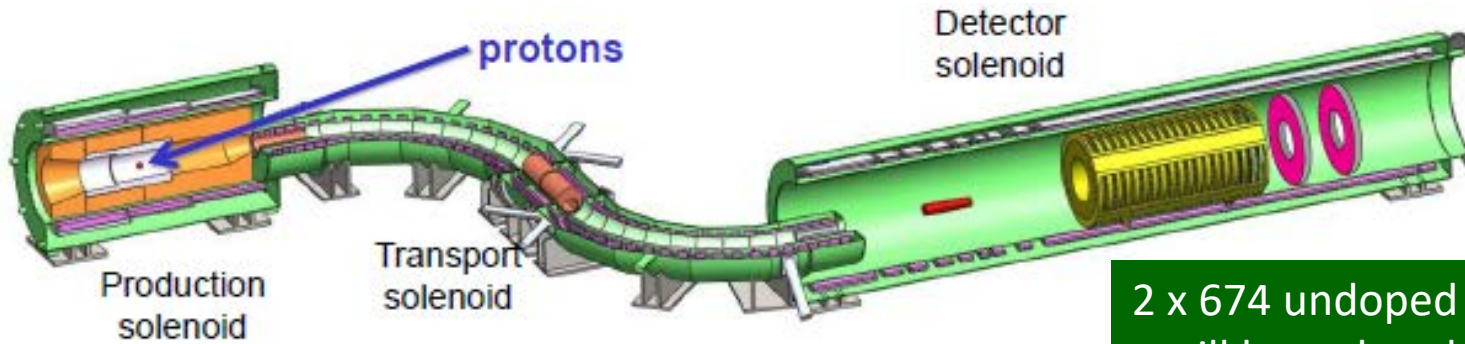
No damage observed in both transmittance and light output after 220 Mrad ionization dose and  $3 \times 10^{14}$  p/cm<sup>2</sup> of 800 MeV



Very promising for a scintillating ceramics based calorimeter  
 Further investigation: slow component and LuAG:Pr



# Mu2e Specifications for Undoped CsI



2 x 674 undoped CsI crystals will be ordered in August

- ❑ 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  $150 \mu\text{m}$  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%**;
  - ❑ 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

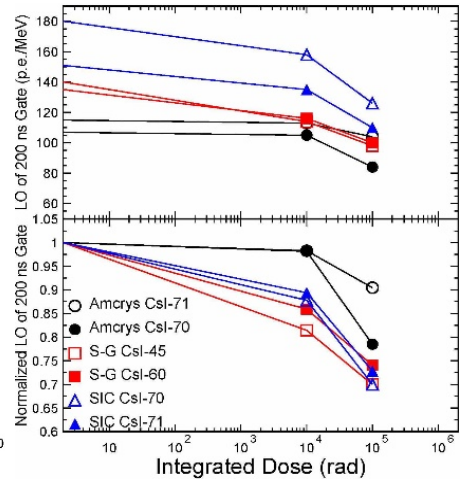
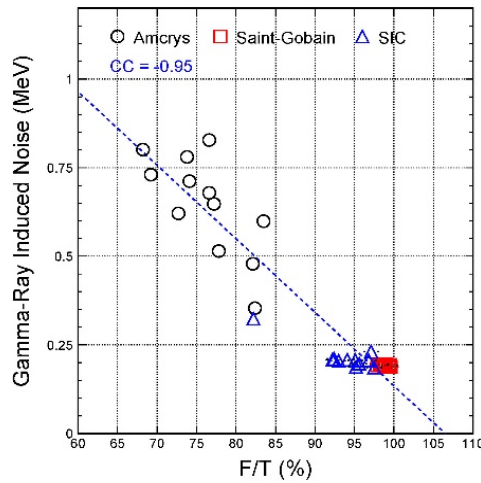
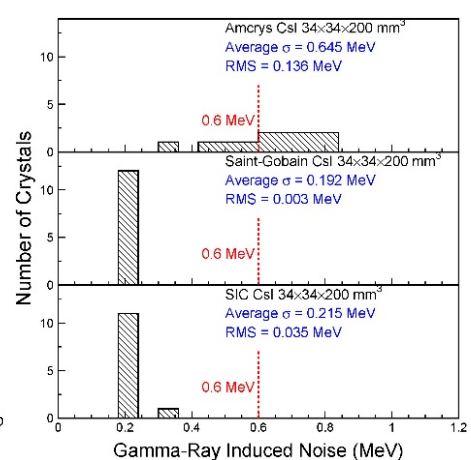
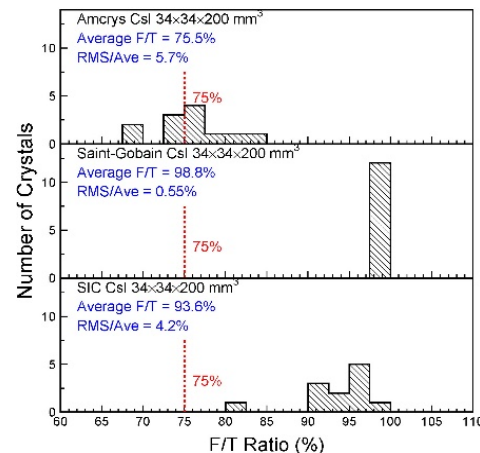
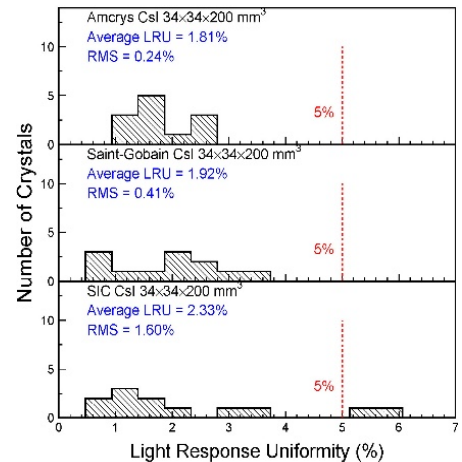
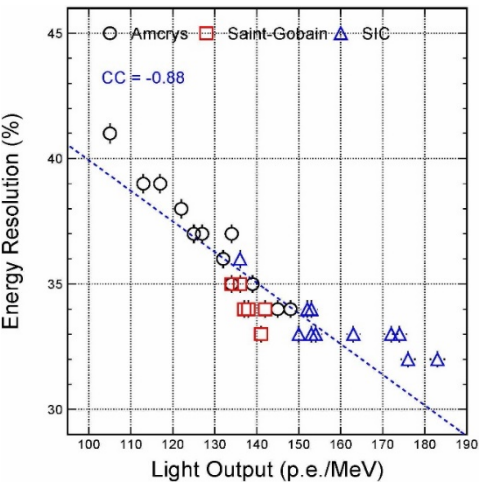
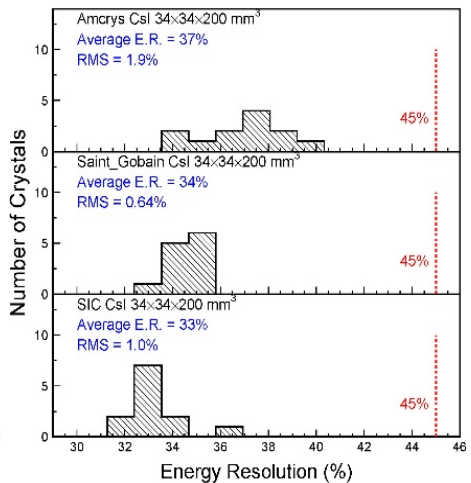
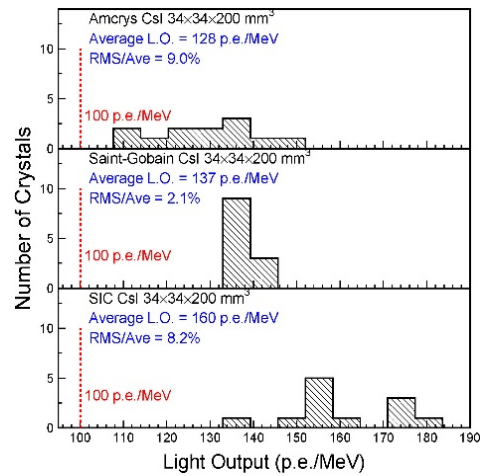
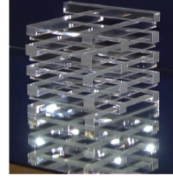


A total of 72 crystals from Amcryst, Saint-Gobain and SICCAS has been measured at Caltech and LNF

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



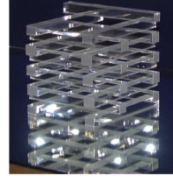
# Quality of Pre-Production CsI



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



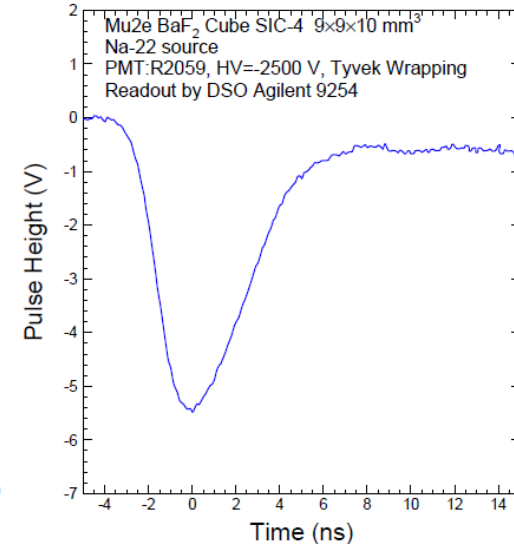
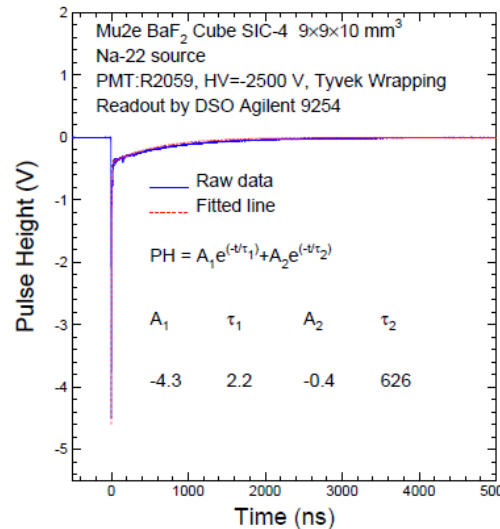
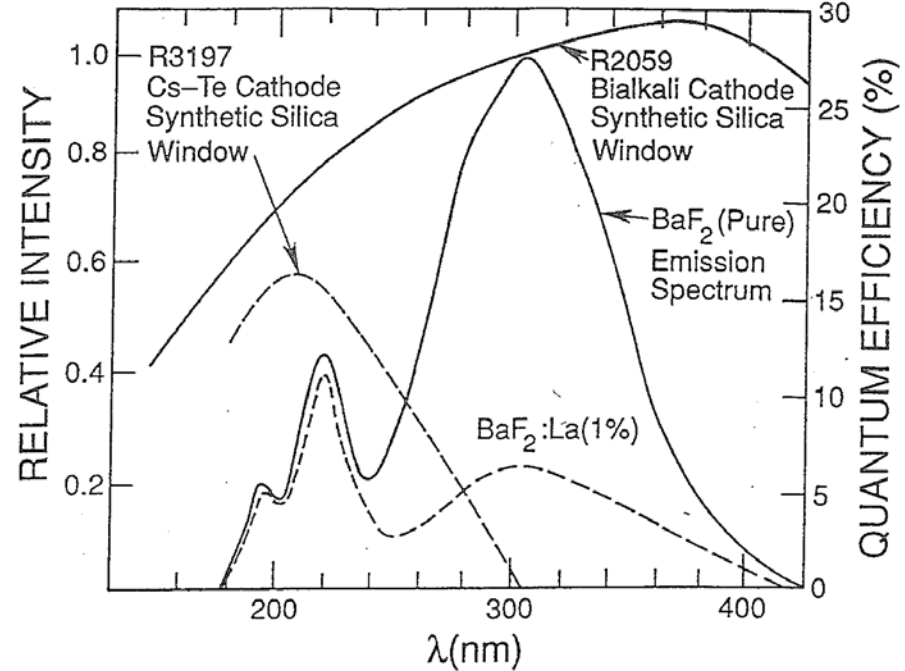
# Fast and Slow Light from BaF<sub>2</sub>



A radiation level exceeding 100 krad is expected at the proposed Mu2e-II, so BaF<sub>2</sub> is being considered.

The amount of light in the fast component of BaF<sub>2</sub> 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.



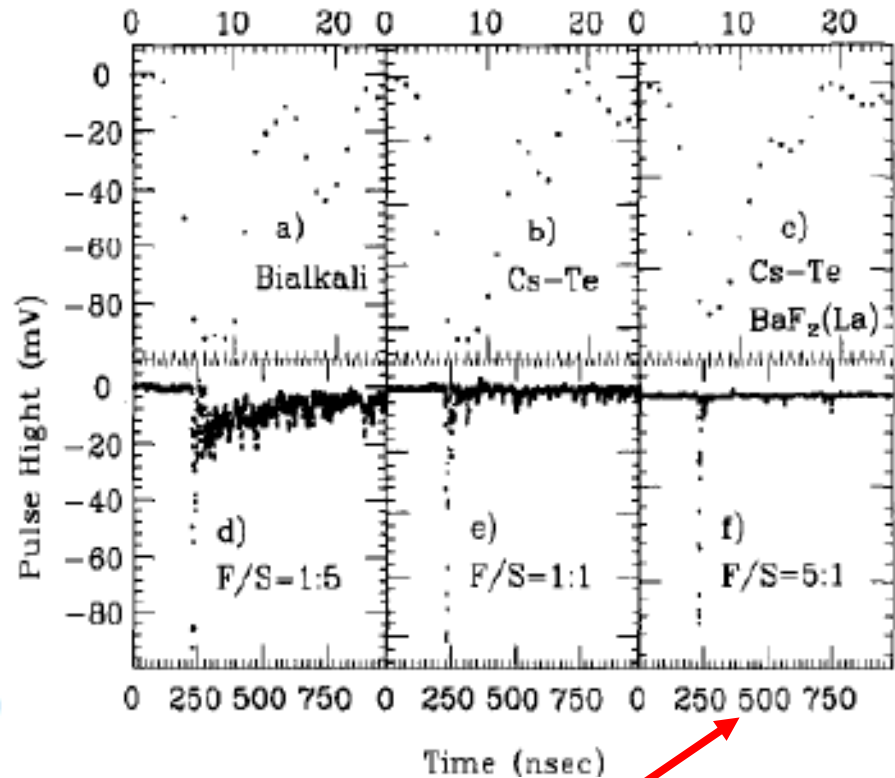
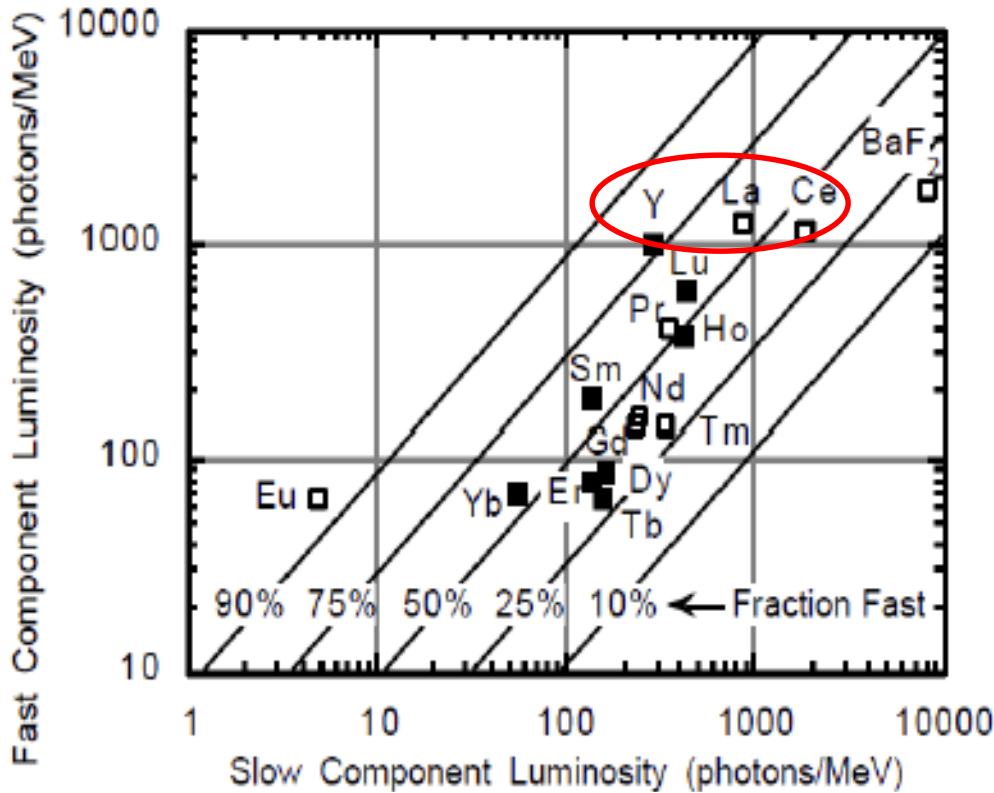


# Slow Suppression: Doping & Readout



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

B.P. SOBOLEV et al., "SUPPRESSION OF BaF<sub>2</sub> SLOW COMPONENT OF X-RAY LUMINESCENCE IN NON-STOICHIOMETRIC Ba<sub>0.9</sub>R<sub>0.1</sub>F<sub>2</sub> CRYSTALS (R=RARE EARTH ELEMENT)," *Proceedings of The Material Research Society: Scintillator and Phosphor Materials*, pp. 277-283, 1994.



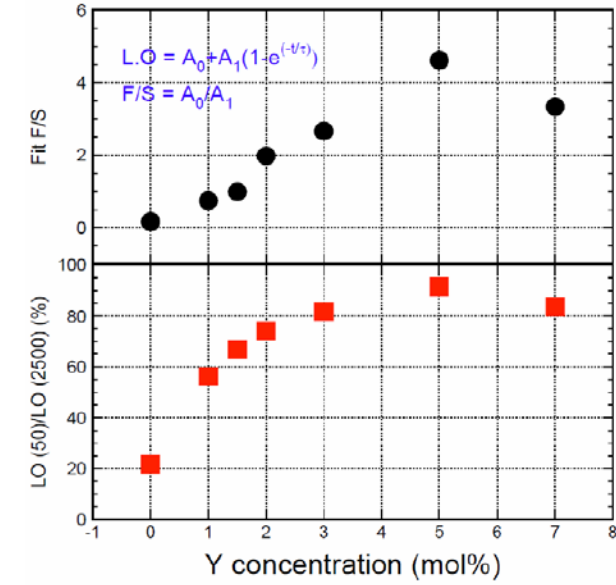
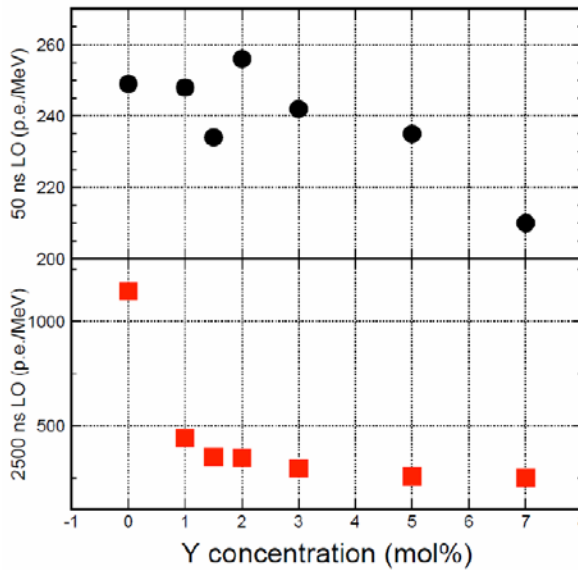
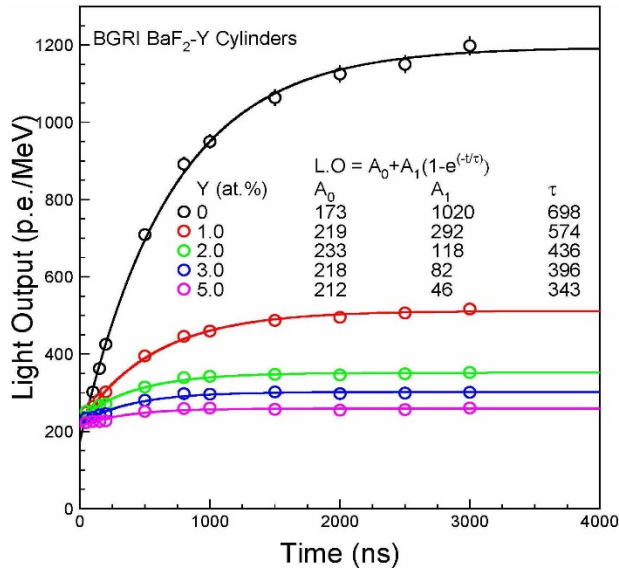
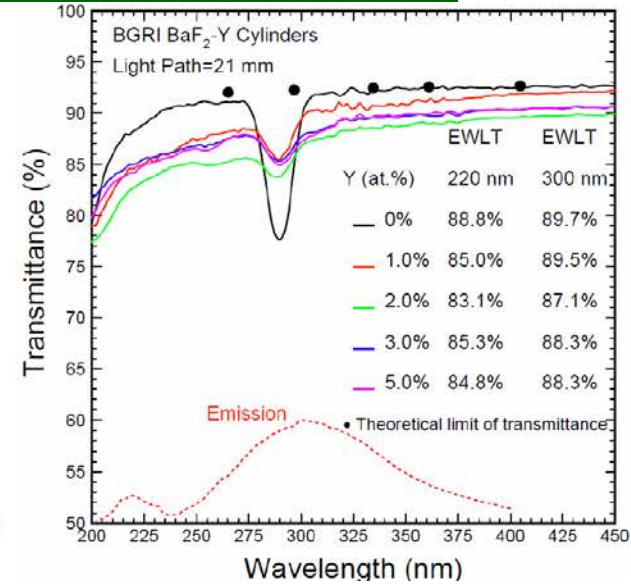
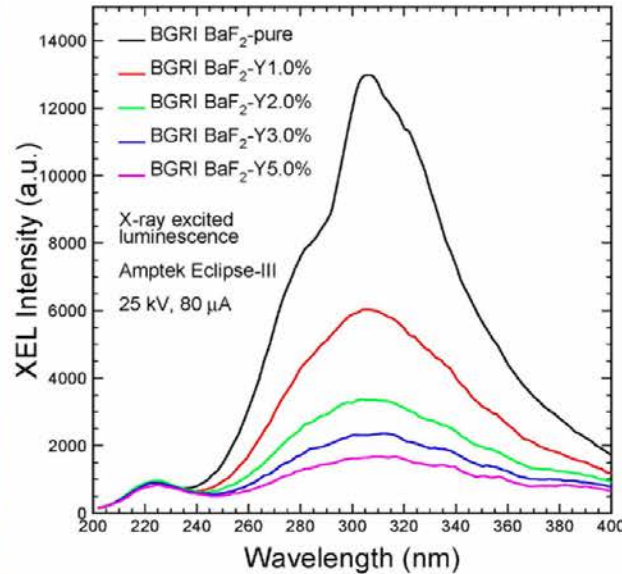
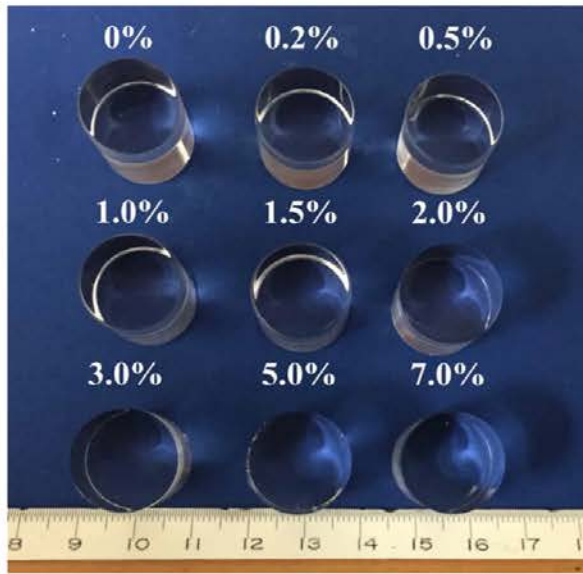
Solar-blind cathode (Cs-Te) + La doping achieved F/S = 5/1



# Yttrium Doping in BaF<sub>2</sub>



F/S ratio from 1/5 to 5/1: very effective slow suppression







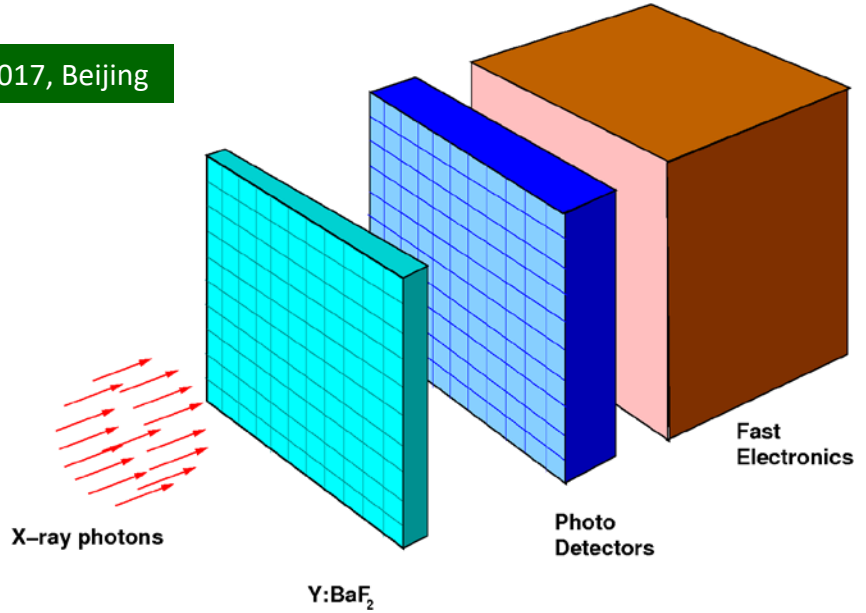
# A Y:BaF<sub>2</sub> Crystal Based Imager



- BaF<sub>2</sub> has good efficiency for hard X-rays.
- Its fast scintillation with sub-ns decay time provides bright light in 2 ns with very little tail.
- Yttrium doping in BaF<sub>2</sub> suppresses its slow scintillation by a factor of 25 and maintains its fast light.

R-Y Zhu, Talk presented in TIPP2017, Beijing

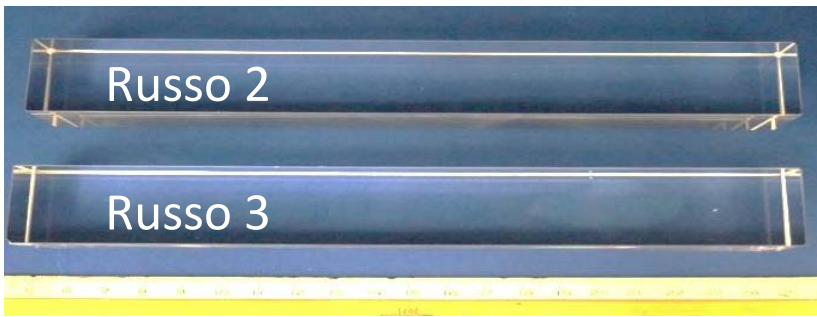
- A detector concept:
  - Pixelized Y:BaF<sub>2</sub> screen;
  - Pixelized fast photodetector;
  - Fast electronics readout.



- To be developed:  
Crystals, DUV photodetectors and readout.



# BGRI/Incrom/SIC BaF<sub>2</sub> Samples



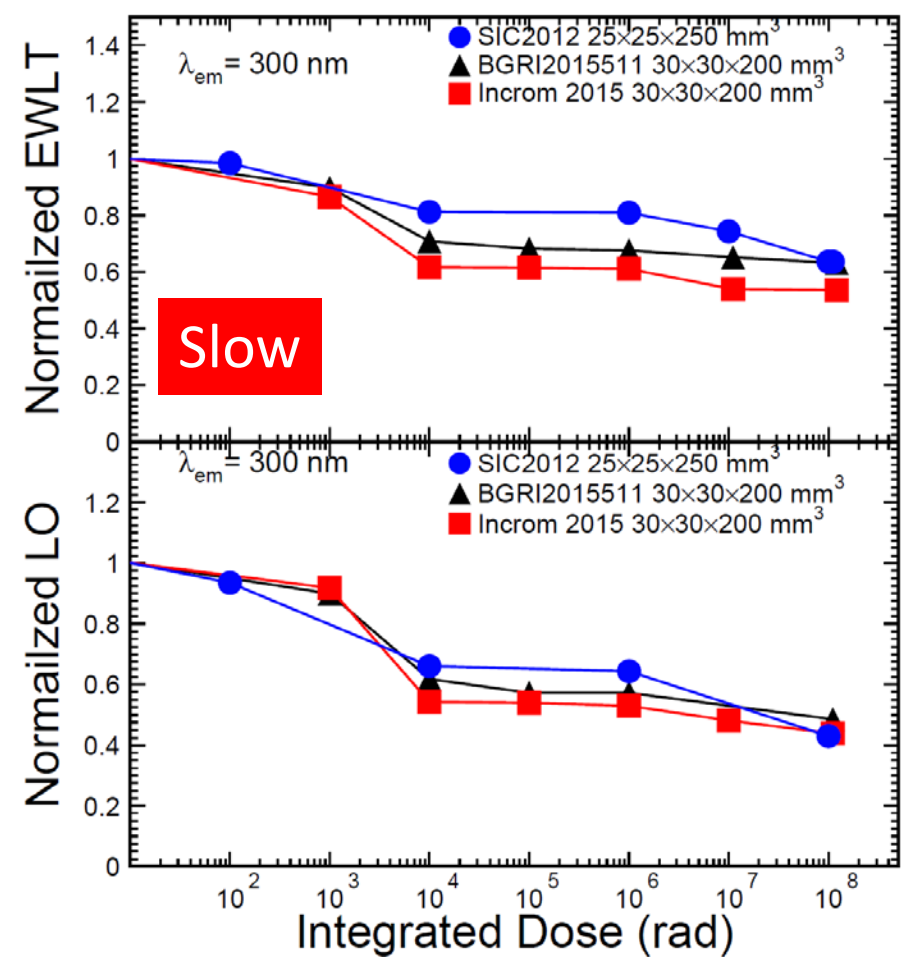
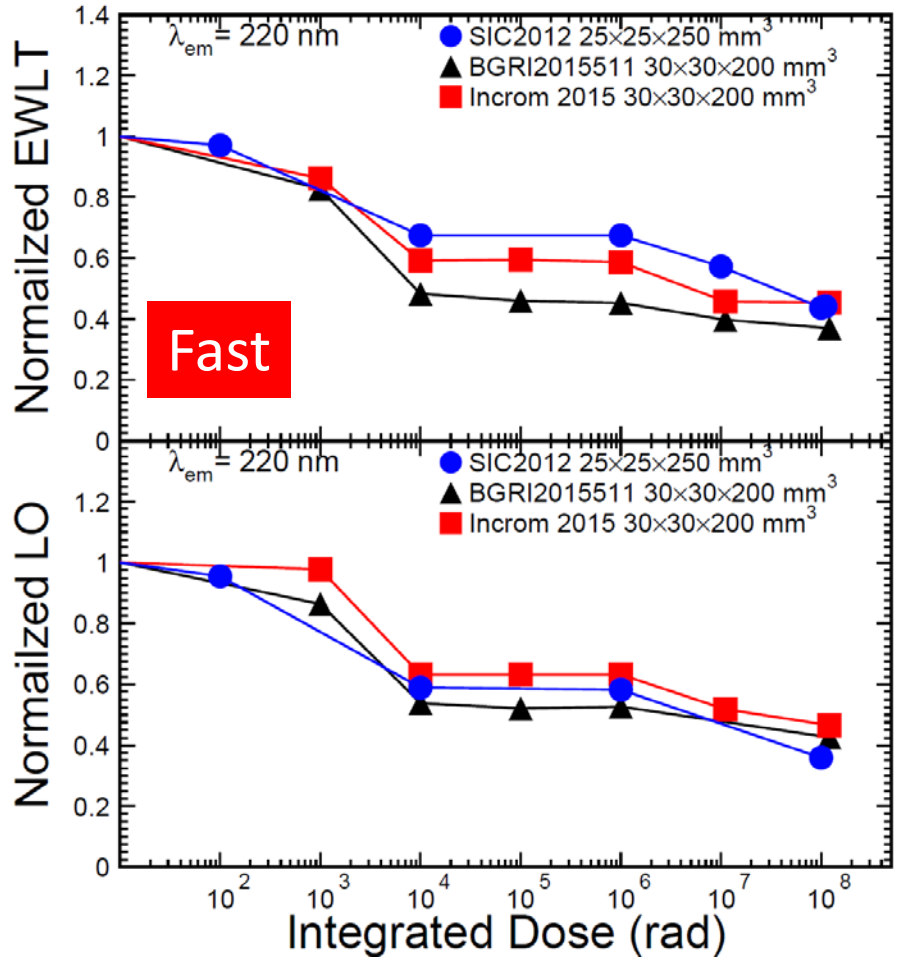
ID	Vendor	Dimension (mm <sup>3</sup> )	Polishing
SIC 1-20	SICCAS	30x30x250	Six faces
BGRI-2015 D, E, 511	BGRI	30x30x200	Six faces
Russo 2, 3	Incrom	30x30x200	Six faces



# BaF<sub>2</sub>: Normalized EWLT and LO



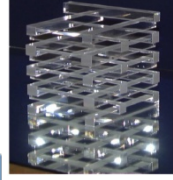
Consistent damage in crystals from three vendors



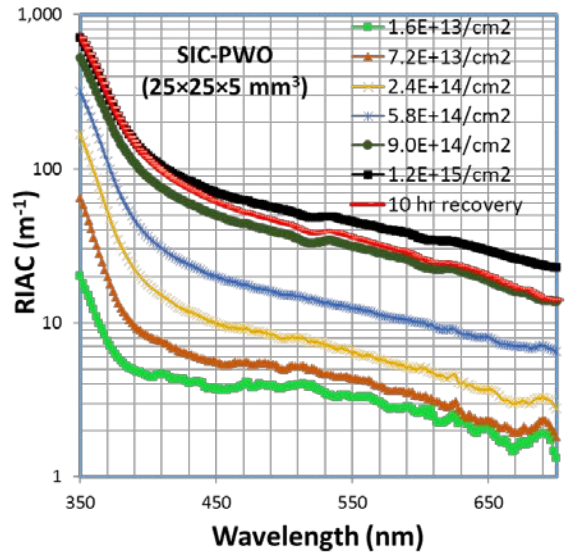
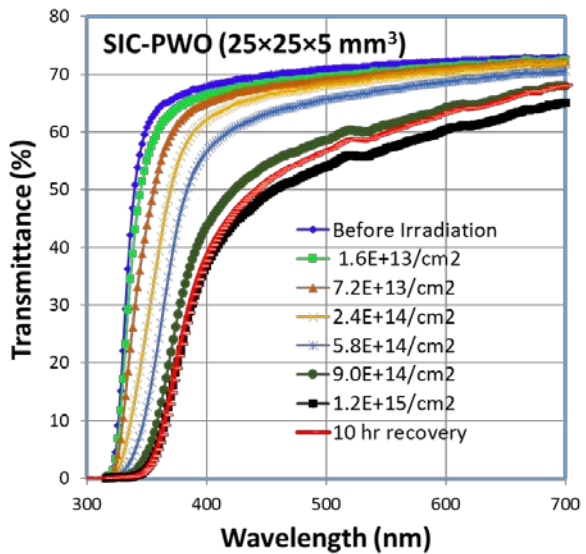
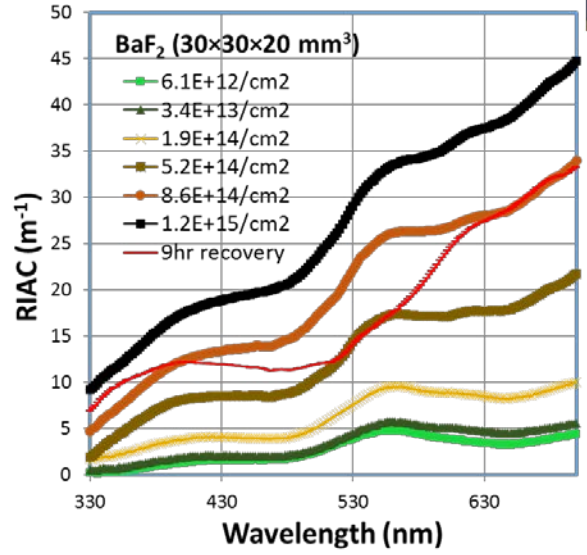
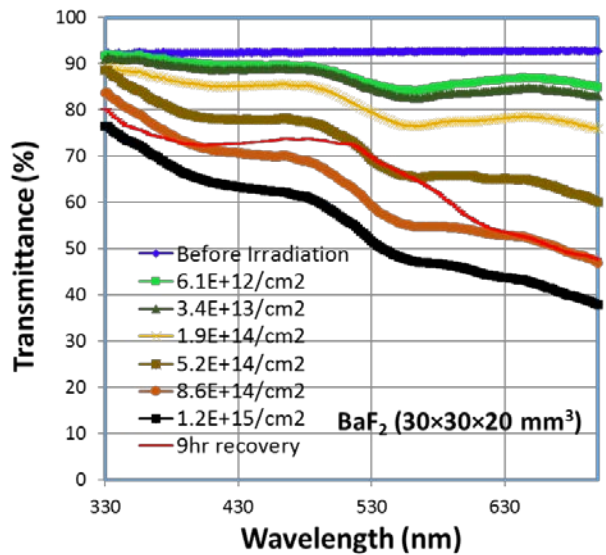
Remaining light output after 120 Mrad: 40%/45% for the fast/slow component



# 800 MeV Proton Damage in BaF<sub>2</sub> & PWO



A Hellma BaF<sub>2</sub> of 2 cm was irradiated from  $6.1 \times 10^{12}$  to  $1.2 \times 10^{15}$  p/cm<sup>2</sup> in six steps with transmittance (330-650 nm) measured *in-situ*. The sample will be measured at Caltech for 200 – 650 nm.

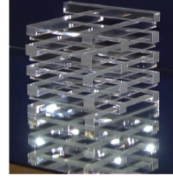


A 5 mm thick SIC PWO plate was irradiated from  $1.6 \times 10^{13}$  to  $1.2 \times 10^{15}$  p/cm<sup>2</sup> with transmittance (300-700 nm) measured *in-situ*. The RIAC at 420 nm was measured to be 13.1 / 92.2 cm<sup>-1</sup> after  $2.4 \times 10^{14}$  /  $1.2 \times 10^{15}$  p/cm<sup>2</sup>.

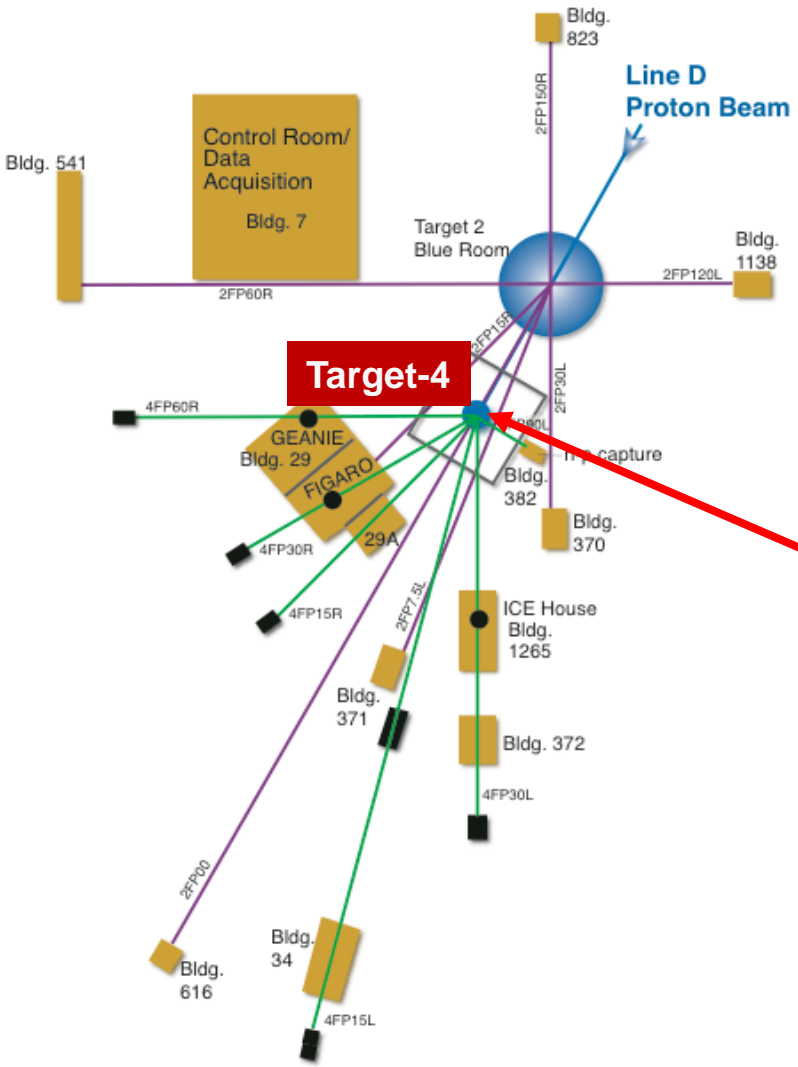
R.-Y. Zhu, "Preliminary Report on the Experiment 7324 with 800 MeV Protons at Los Alamos," [http://www.hep.caltech.edu/~zhu/talks/ryz\\_1601207\\_LANL.pdf](http://www.hep.caltech.edu/~zhu/talks/ryz_1601207_LANL.pdf)



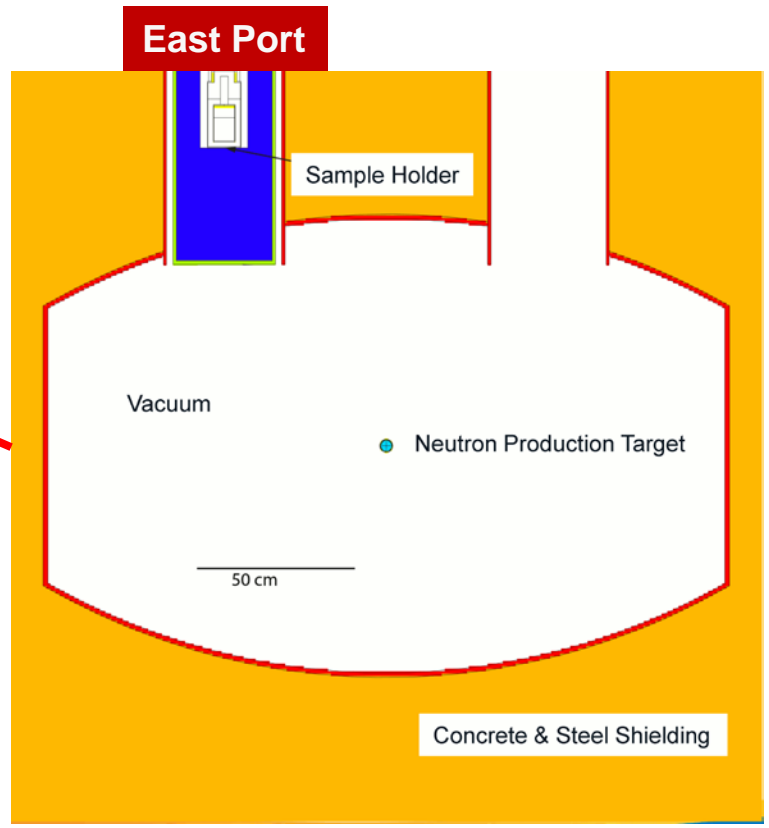
# Neutron Irradiation Test at LANL



## Los Alamos Neutron Science Center (LANSCE)



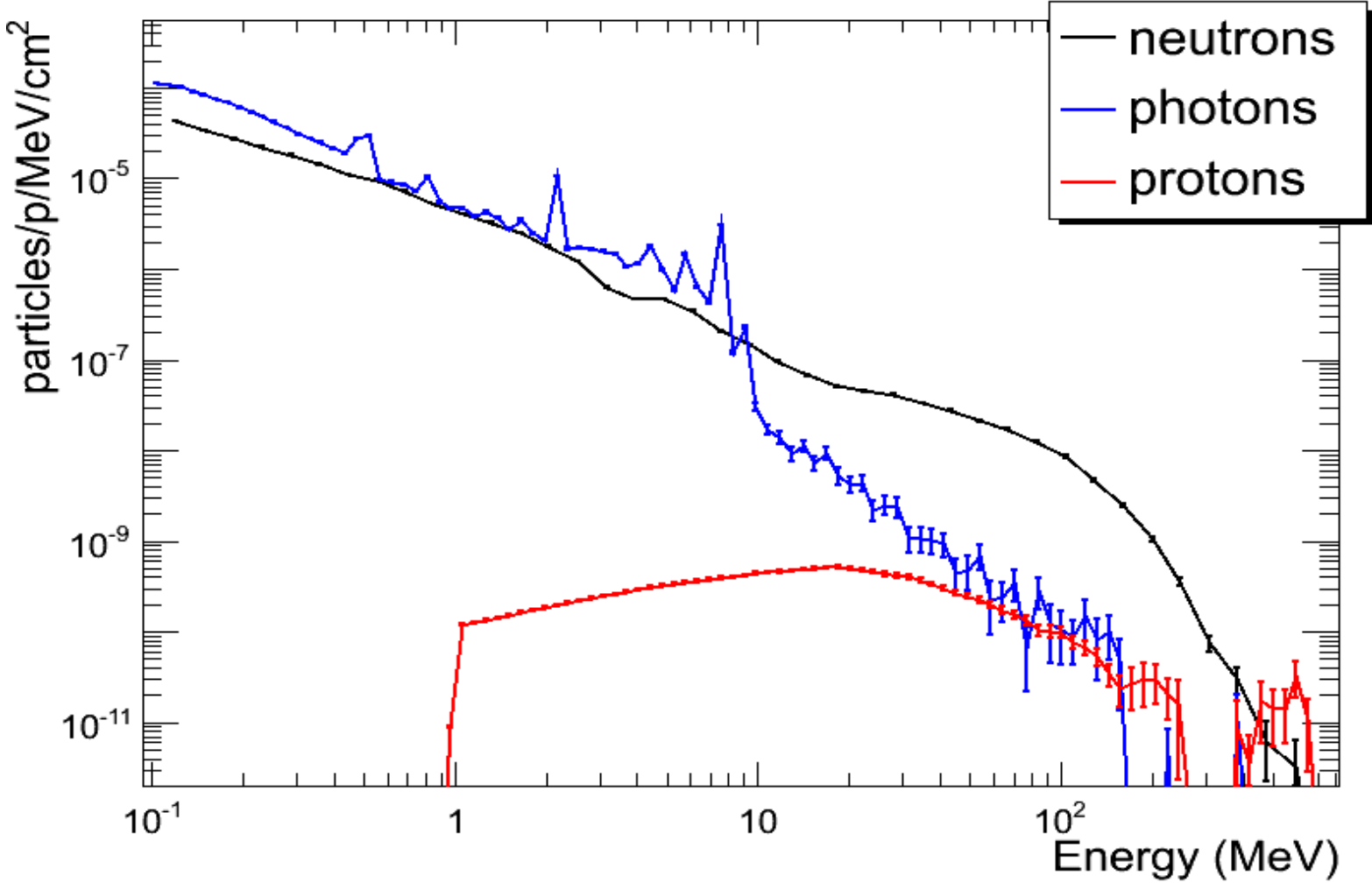
Samples are placed at Target-4 East Port, about 1.2 m away from the neutron production target.





# 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)





# Estimated Fluence and Dose



3 groups of samples were irradiated for 21.2, 46.3 and 119.8 days respectively with the neutron and proton fluences and ionization dose calculated by using the 800 MeV proton beam data.

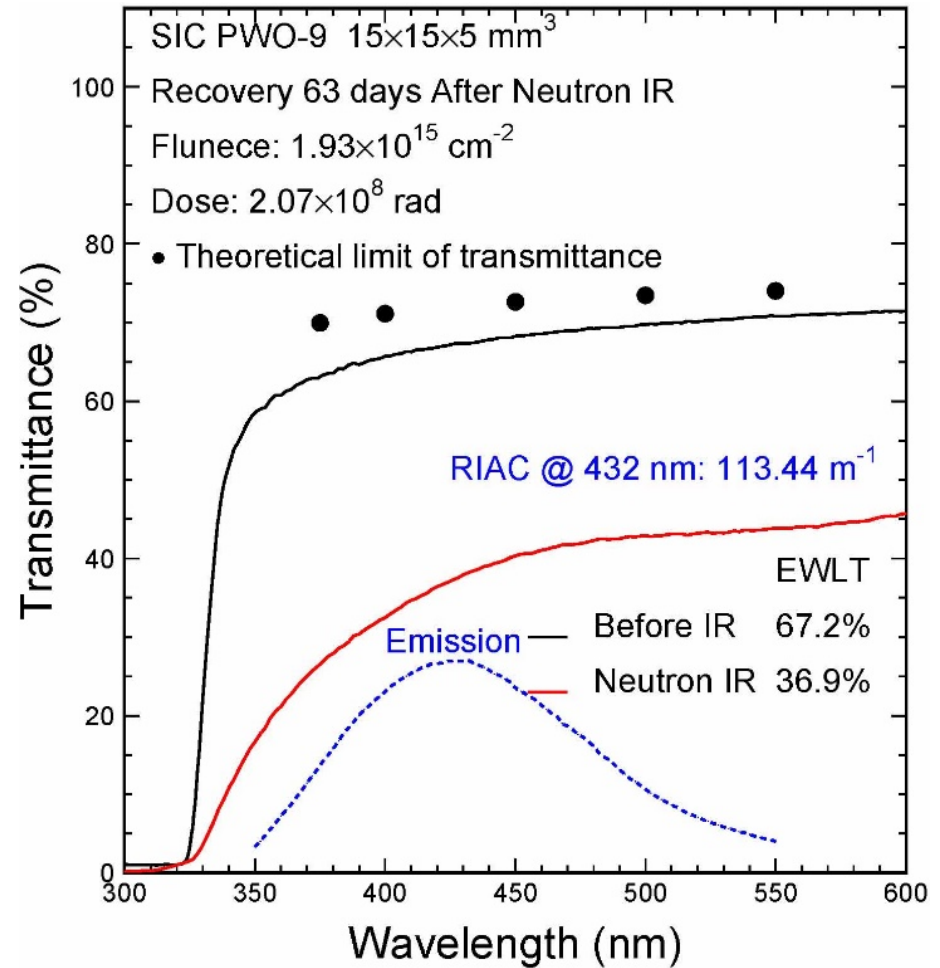
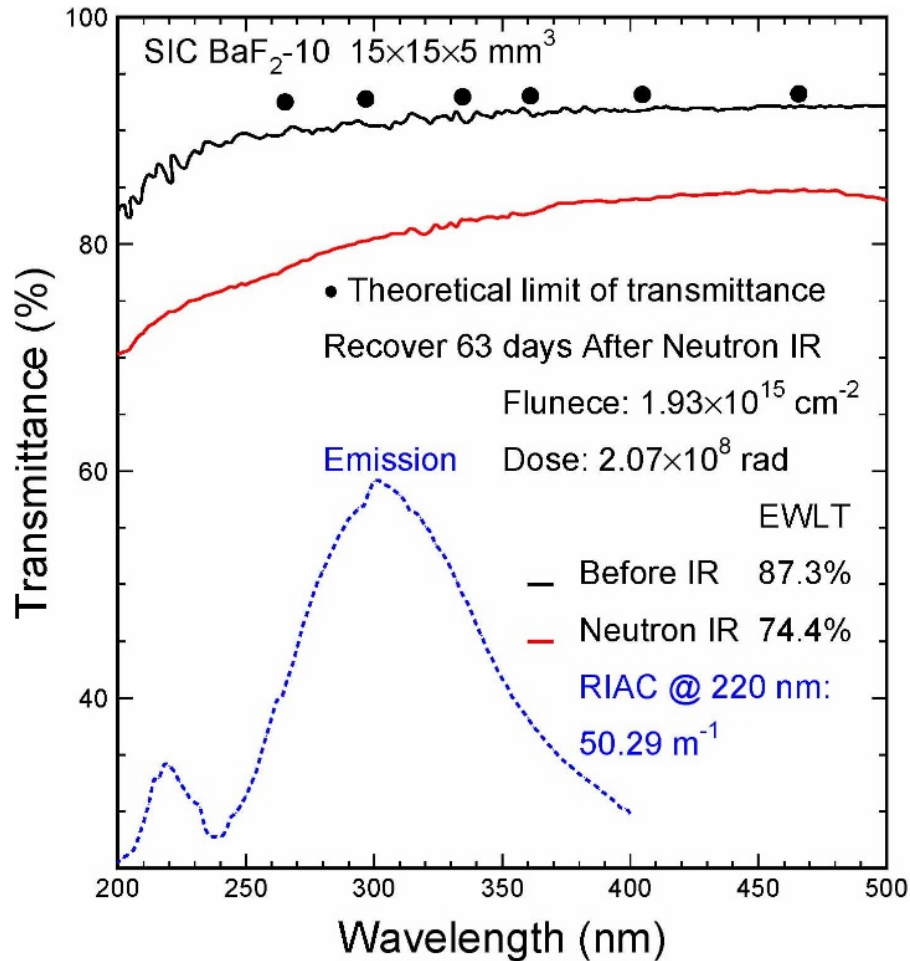
Particles	Group-1 Fluence (cm <sup>-2</sup> )	Group-2 Fluence (cm <sup>-2</sup> )	Group-3 Fluence (cm <sup>-2</sup> )
Thermal and Epithermal Neutrons (0<En < 1 eV)	1.23E+15	2.69E+15	6.04E+15
Slow and Intermediate Neutrons (1 eV <En < 1 MeV)	4.50E+15	9.80E+15	2.20E+16
<b>Fast neutrons Fluence 1: (En &gt; 1 MeV)</b>	<b>3.94E+14</b>	<b>8.58E+14</b>	<b>1.93E+15</b>
<b>Fast neutrons Fluence 2: (En&gt;20 MeV)</b>	<b>7.64E+13</b>	<b>1.66E+14</b>	<b>3.74E+14</b>
Protons (Ep>1 MeV)	9.34E+11	2.03E+12	4.57E+12
Protons Dose (rad)	2.44E+04	5.32E+04	1.20E+05
Photons (Eg>150 KeV)	1.18E+15	2.57E+15	5.78E+15
<b>Photons Dose (rad)</b>	<b>4.22E+07</b>	<b>9.21E+07</b>	<b>2.07E+08</b>
<b>Photons Dose (rad) with 5 mm Pb shielding</b>	<b>3.00E+07</b>	<b>6.54E+07</b>	<b>1.47E+08</b>



# Transmittance Damage in BaF<sub>2</sub> & PWO



Samples in three groups irradiated at the East Port of LANSCE



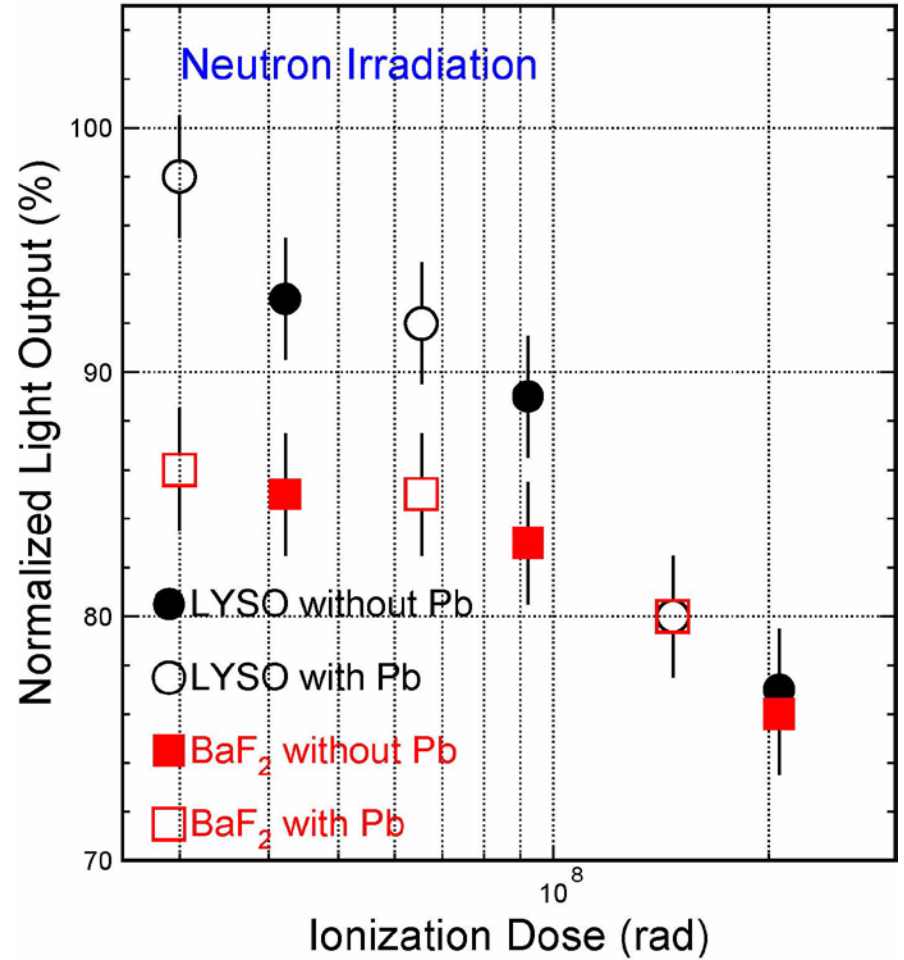
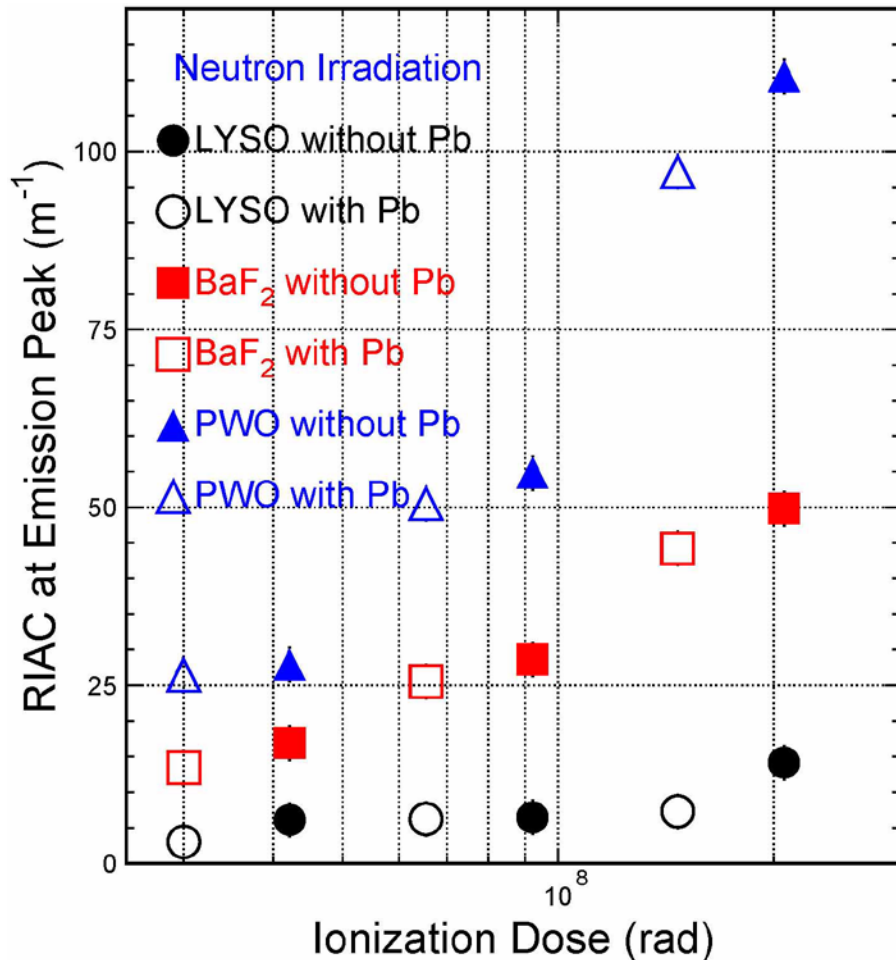




# RIAC & LO Vs. Ionization Dose



Results consist with ionization dose induced damage, indicating no neutron specific damage in LYSO, BaF<sub>2</sub> & PWO





# Summary



- ❑ **LYSO crystals and LuAG ceramics are robust scintillators against ionization dose as well as charged and neutral hadrons.**
- ❑ **Commercially available undoped BaF<sub>2</sub> crystals provide sufficient fast light with sub-ns decay time and excellent radiation hardness beyond 100 Mrad and  $1 \times 10^{15}$  p/cm<sup>2</sup>. They promise a very fast and robust calorimeter in a severe radiation environment.**
- ❑ **Without using a selected readout yttrium doping in BaF<sub>2</sub> crystals increases the F/S ratio from 1/5 to 5/1 while keeping the intensity of the sub-ns fast component unchanged. The slow contamination at this level is already much less than commercially available undoped CsI.**
- ❑ **Results of the experiments 6991 and 7332 at LANL show fast neutrons up to  $2 \times 10^{15}$  n/cm<sup>2</sup> do not damage LYSO, BaF<sub>2</sub> and PWO crystals.**
- ❑ **Our plan is to investigate LYSO:Ce,Ca crystals, LuAG:Ce and LuAG:Pr ceramics, and radiation hardness of Y:BaF<sub>2</sub> crystals. Will also pay an attention to photodetector with DUV response: LAPPD, Si or diamond based solid state detectors.**



# Diamond Photodetector



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

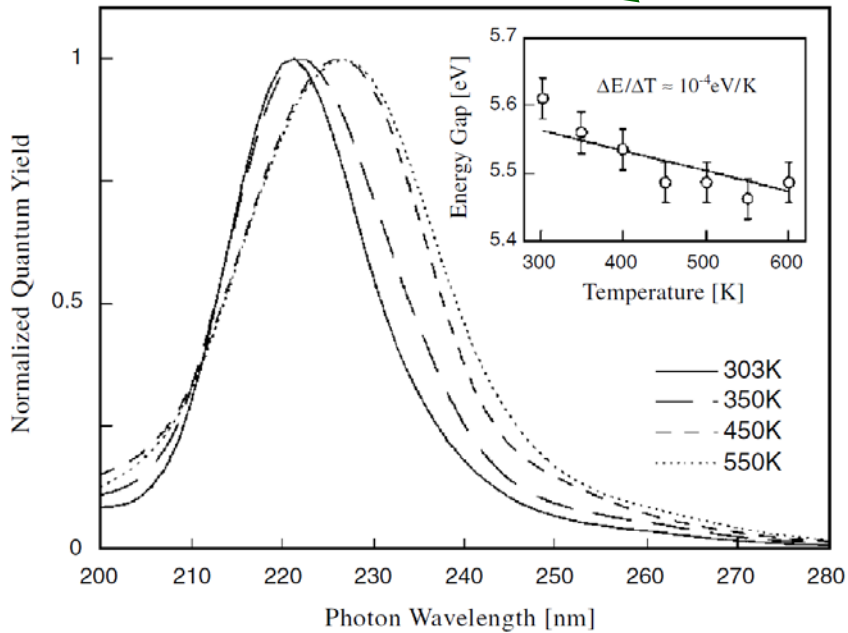


Figure 6. Quantum efficiency of diamond photoconductors at different temperatures and Arrhenius plot of the peak value (inset). (From [Sal00].)

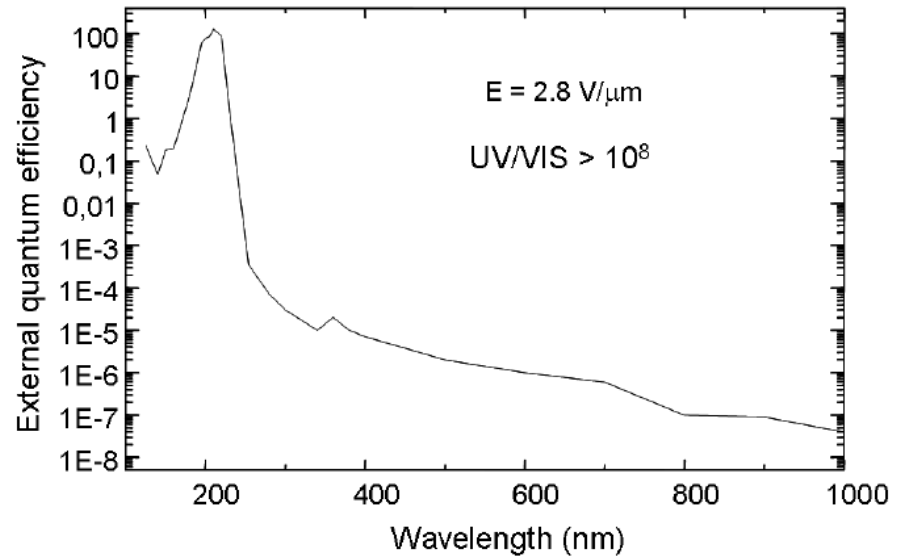
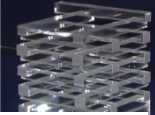


Fig. 4. External quantum efficiency extended to visible and near infrared wavelength regions. The

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

# GHz Hard X-Ray Imagers



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

Organizers: Peter Denes, Sol Gruner, Michael Stevens & Zhehui (Jeff) Wang<sup>1</sup>  
 (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	$\leq 300 \mu\text{m}$	$< 300 \mu\text{m}$
Dynamic range	$10^3$ X-ray photons	$\geq 10^4$ 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**