Development of Transparent Ceramic and Glass Scintillators for Future HEP Applications

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2019 DOE Basic Research Needs Study on Instrumentation: Calorimetry

Priority Research Direction
PRD 1: Enhance calorimetry energy resolution for precision electroweak mass and missing-energy measurements
PRD 2: Advance calorimetry with spatial and timing resolution and radiation hardness to master high-rate environments
PRD 3: Develop ultrafast media to improve background rejection in calorimeters and improve particle identification

Fast/ultrafast, radiation hard and cost-effective inorganic scintillators are needed to achieve energy, spatial and timing resolutions required by BRN

Since 1928

Fast and Ultrafast Inorganic Scintillators

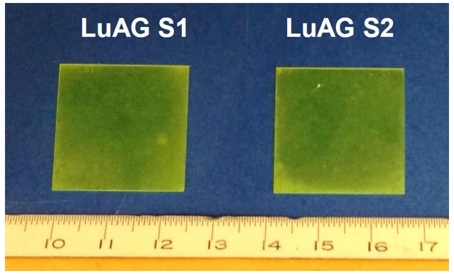
See in **Snowmass Whitepaper**, arXiv: 2203.06788

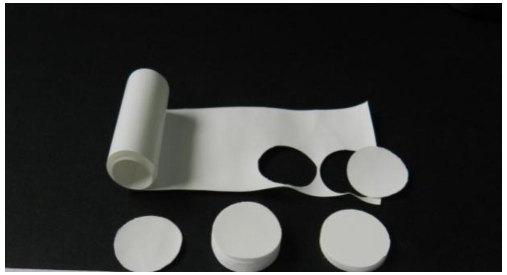
	BaF_2	BaF_2:Y	ZnO:Ga	Lu_2O_3:Yb	YAP:Yb	YAG:Yb	β-Ga_2O_3	PWO	LYSO:Ce	LuAG:Ce	YAP:Ce	GAGG:Ce	LuYAP:Ce	YSO:Ce
Density (g/cm^3)	4.89	4.89	5.67	9.42	5.35	4.56	5.94	8.28	7.4	6.76	5.35	6.5	7.2[f]	4.44
Melting points (^oC)	1280	1280	1975	2490	1870	1940	1725	1123	2050	2060	1870	1850	1930	2070
X_0 (cm)	2.03	2.03	2.51	0.81	2.59	3.53	2.51	0.89	1.14	1.45	2.59	1.63	1.37	3.10
R_M (cm)	3.1	3.1	2.28	1.72	2.45	2.76	2.20	2.00	2.07	2.15	2.45	2.20	2.01	2.93
λ_I (cm)	30.7	30.7	22.2	18.1	23.1	25.2	20.9	20.7	20.9	20.6	23.1	21.5	19.5	27.8
Z_{eff}	51.0	51.0	27.7	67.3	32.8	29.3	27.8	73.6	63.7	58.7	32.8	50.6	57.1	32.8
dE/dX (MeV/cm)	6.52	6.52	8.34	11.6	7.91	7.01	8.82	10.1	9.55	9.22	7.91	8.96	9.82	6.57
λ_{peak} [a] (nm)	300 220	300 220	380	370	350	350	380	425 420	420	520	370	540	385	420
Refractive Index[b]	1.50	1.50	2.1	2.0	1.96	1.87	1.97	2.20	1.82	1.84	1.96	1.92	1.94	1.78
Normalized Light Yield[a,c]	42 4.8	1.7 4.8	6.6[d]	0.95	0.19[d]	0.36[d]	6.5 0.5	1.6 0.4	100	35[e] 48[e]	9 32	190	16 15	80
Total Light yield (ph/MeV)	13,000	2,000	2,000[d]	320	57[d]	110[d]	2,100	130	30,000	25,000[e]	12,000	58,000	10,000	24,000
Decay time[a] (ns)	600 0.5	600 0.5	<1	1.1[d]	1.5	4	148 6	30 10	40	820 50	191 25	570 130	1485 36	75
LY in 1st ns (photons/MeV)	1200	1200	610[d]	190	28[d]	24[d]	43	5.3	740	240	391	400	125	318
LY in 1st ns /Total LY (%)	9.2	60	31	61	49	22	2.0	4.3	2.5	1.0	3.3	0.7	1.3	1.3
40 keV Att. Leng. (1/e, mm)	0.106	0.106	0.407	0.127	0.314	0.439	0.394	0.111	0.185	0.251	0.314	0.319	0.214	0.334

[a] top/bottom row: slow/fast component; [b] at the emission peak; [c] normalized to LYSO:Ce; [d] excited by Alpha particles; [e] 0.3 Mg at% co-doping; [f] $Lu_{0.7}Y_{0.3}AlO_3$:Ce.

Why Scintillating Ceramics

- **Ceramics provide a cost-effective solution for future HEP experiments.**
  - **Simple production technology;**
  - **High raw material usage;**
  - **Minimum after-growth mechanical processing.**





- **Unlike single crystal, ceramic fabrication does not require melting raw material.**
  - **Lower sintering temperature;**
  - **Dopants distribute homogeneously without segregation process;**
  - **Can be made into complex structure.**

Cubic Structure Ceramics can be Transparent

Material	Form[a]	Crystal system[b]	Transparency	Density (g/cm^3)
$Y_{1.34}Gd_{0.6}Eu_{0.06}O_3$	C	C	Transparent	5.92
Gd_2O_2S:Pr,Ce,F	C	H	Translucent	7.34
$Gd_3Ga_5O_{12}$:Cr,Ce	C	C	Transparent	7.09
$BaHfO_3$:Ce	C	C	Opaque	8.35

Crystal system: C = cubic; H = hexagonal; M = monoclinic.

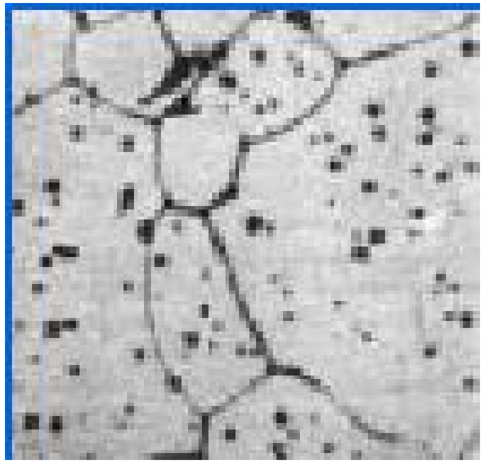
Annu. Rev. Mater. Sci. 1997. 27:69–88

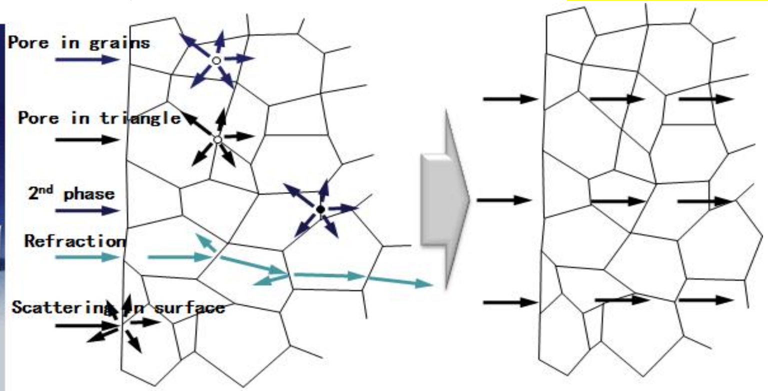
Cost-effective transparent ceramics are pursued by industry

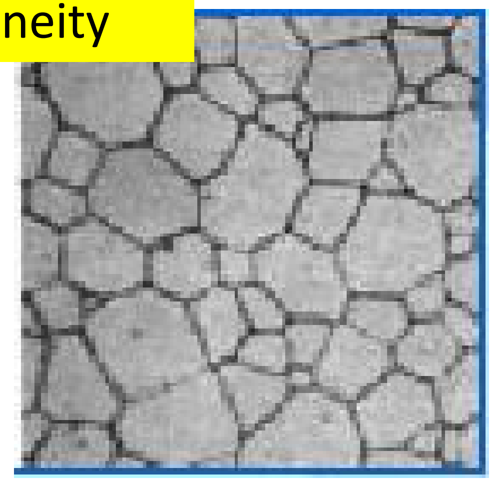
Incident light

Anisotropy

Homogeneity







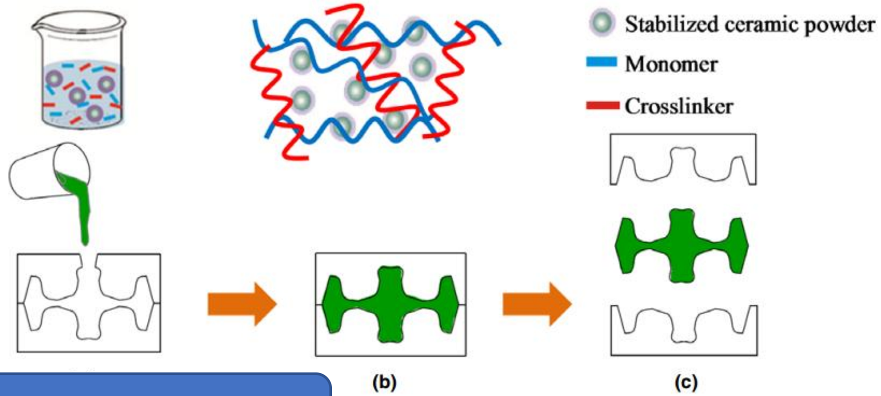
Opaque or translucent

Transparent

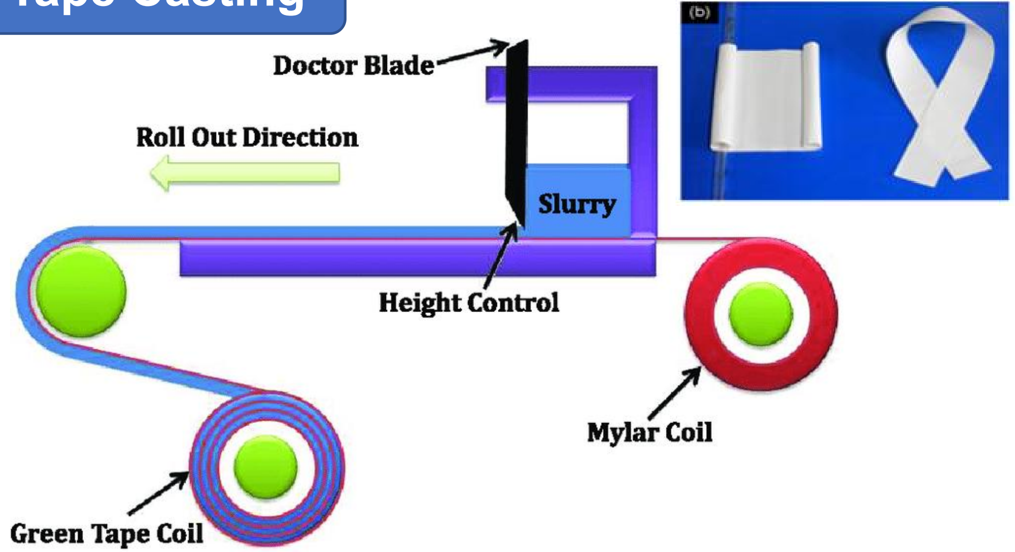
Ceramic Fabrication Process

Ceramic State	Processing Step	Step Objective
Powder	Calcination & Ball milling	Manufacture ceramic powder of required composition and size.
Dry powder or slurry	Mixing & Milling	Homogenous mixture of powder, binder, and additives (and suspension medium in the case of wet forming).
Green body	**Forming**	Shape component into desired geometry.
Brown body	Debinding	Binder burnout and pre-consolidation.
Dense sintered body	Furnace sintering	Densification of the polycrystalline ceramic part with desired microstructure.
Final part	Machining	Dimensions and surface finish within tolerances.

Slip Casting



Tape Casting



Complex structure (rod, dome, sandwich etc.) can be fabricated with minimum after-growth processing

Background: Lu_2O_3:Yb Ceramics

2005—1st report on Lu_2O_3:Yb ceramics

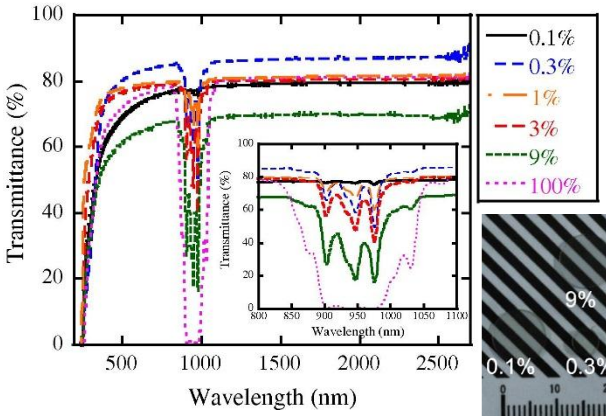
- Takaichi K et al., *phys. stat. sol.* (a) 202, R1-R3 (2005)

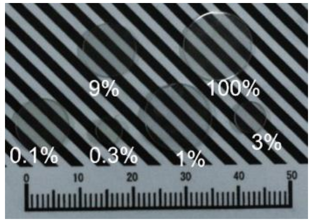
2011—Lu_2O_3:Yb ceramics fabricated by hot-pressing method

- Sanghera J et al., *Opt. Mater.* 33, 670-674 (2011)

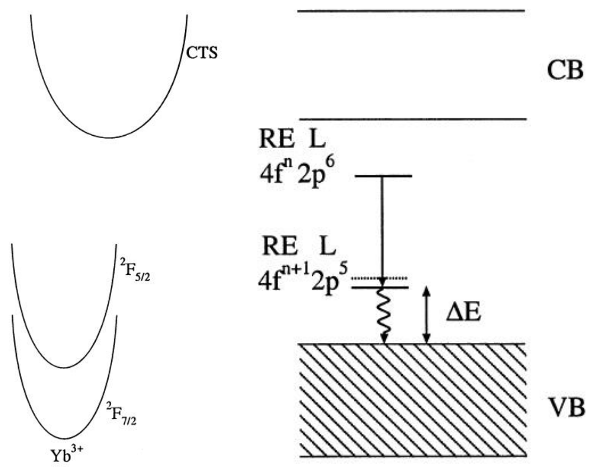
2014—Lu_2O_3:Yb ceramics as a heavy and ultrafast scintillator

- Yanagida T et al., *Opt. Mater.* 36, 1044-1048 (2014)



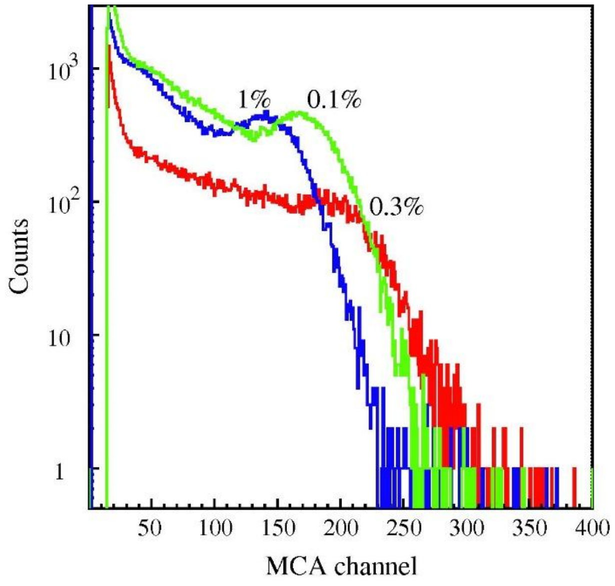


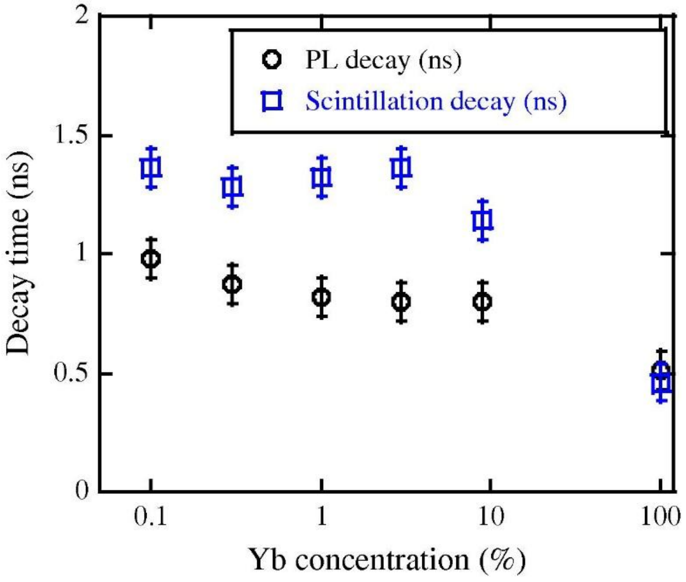
Excellent optical quality approaches theoretical transmittance observed in Lu_2O_3:Yb ceramic samples



Yb^{3+} charge transfer luminescence and thermal quenching mechanism

van Pietersen L et al., J. Lumin. 91, 177-193 (2000)





Because of the thermal quenching nature of Yb^{3+} and CT luminescence light yield of 500 ph/MeV and decay time of ~1 ns were observed

RMD Lu_2O_3:Yb Ceramic Samples

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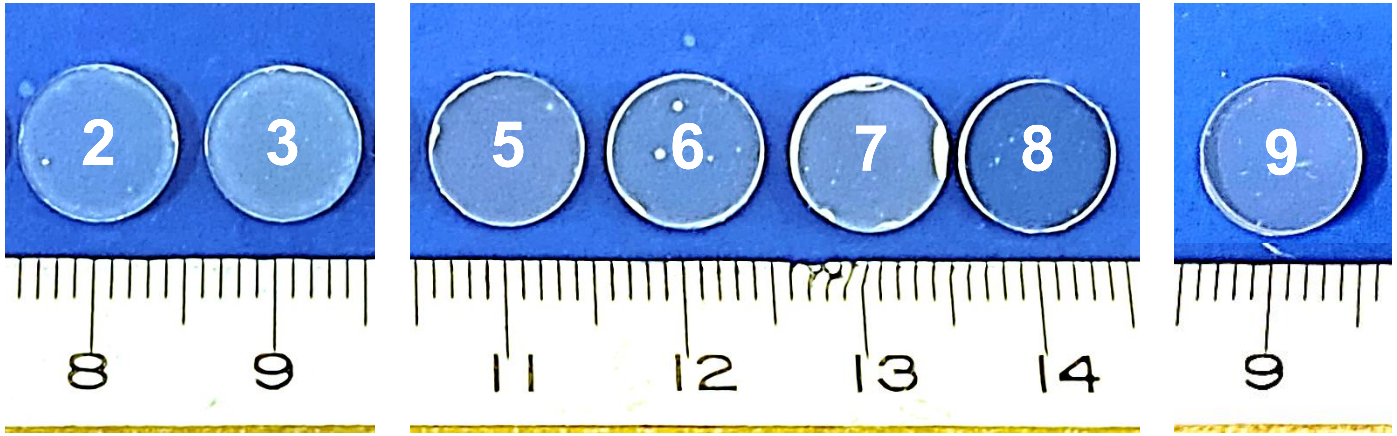

ID	Dimension (mm^3)	Composition
RMD-2	Φ9×1.5	Lu_2O_3
RMD-3	Φ9×1	Lu_2O_3
RMD-5	Φ9×1.5	$(Lu,Y)_2O_3$
RMD-6	Φ9×1.5	$(Lu,Y)_2O_3$
RMD-7	Φ9×2	$(Lu,Y)_2O_3$
RMD-8	Φ9×1	Lu_2O_3
RMD-9	Φ9×2	$(Lu,Y)_2O_3$

	Lu_2O_3	LYSO	BaF_2	LuAG
Density (g/cm^3)	**9.42**	7.4	4.89	6.76
Melting points (°C)	2490	2050	1280	2060
X_0 (cm)	0.81	1.14	2.03	1.45
R_M (cm)	1.72	2.07	3.1	2.15
λ_I (cm)	18.1	20.9	30.7	20.6
Z_{eff}	68.0	64.8	51.6	60.3
dE/dX (MeV/cm)	11.6	9.55	6.52	9.22

Lu_2O_3:Yb is attractive to the HEP community: high density, ultrafast decay and large dE/dX. Single crystal growth is an expensive process due to its very high melting point. Ceramics are a promising approach.

Emission and Transmittance

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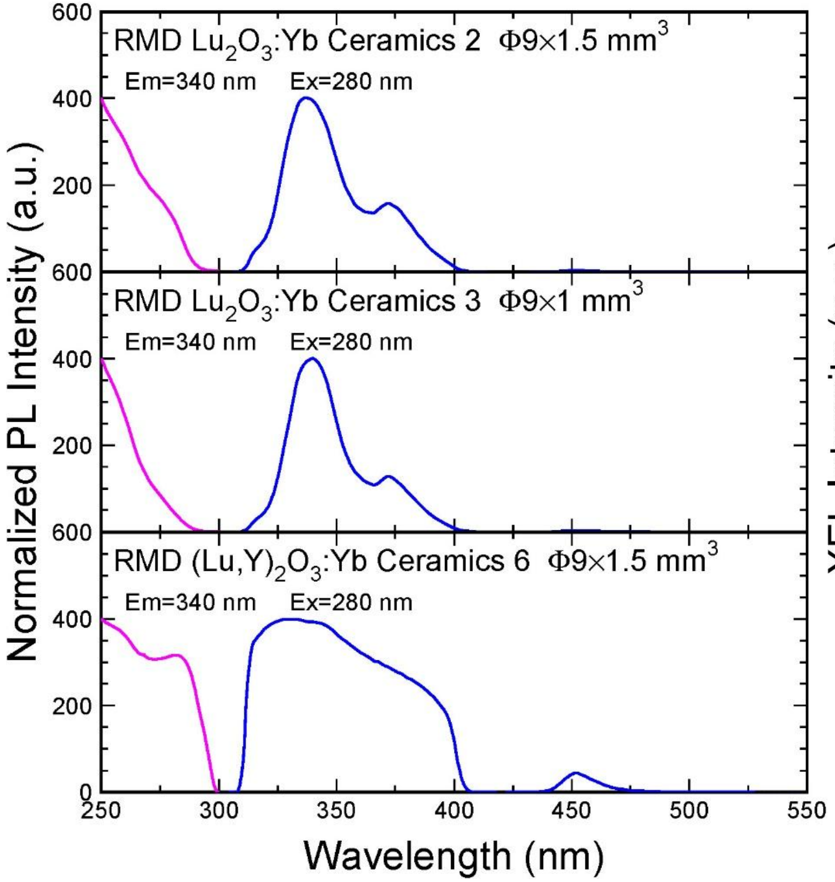
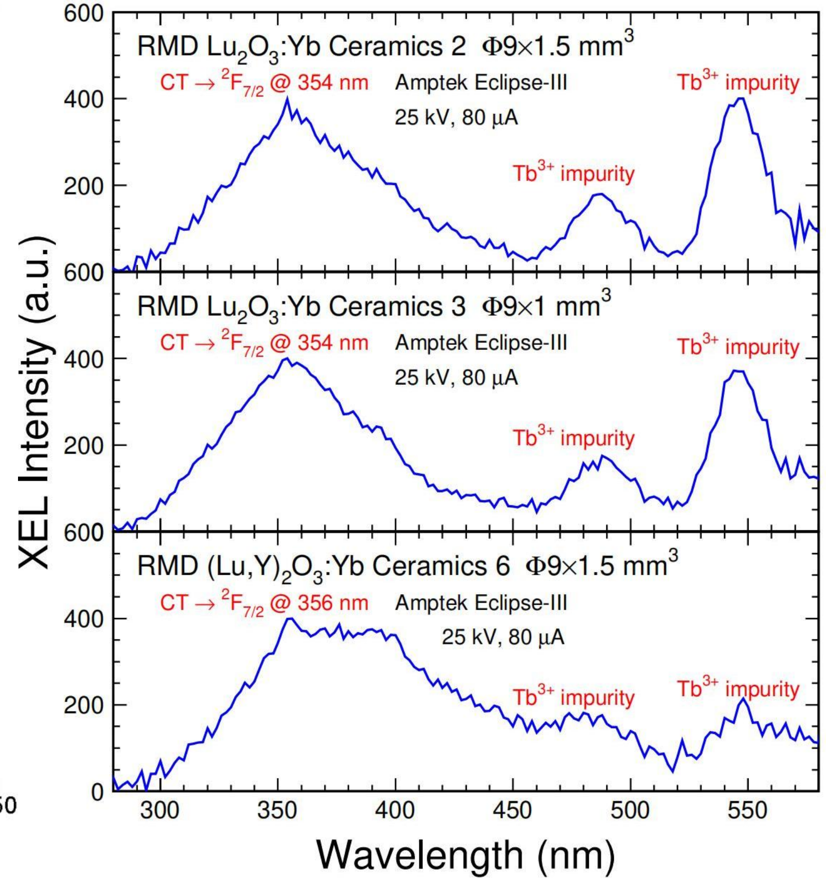
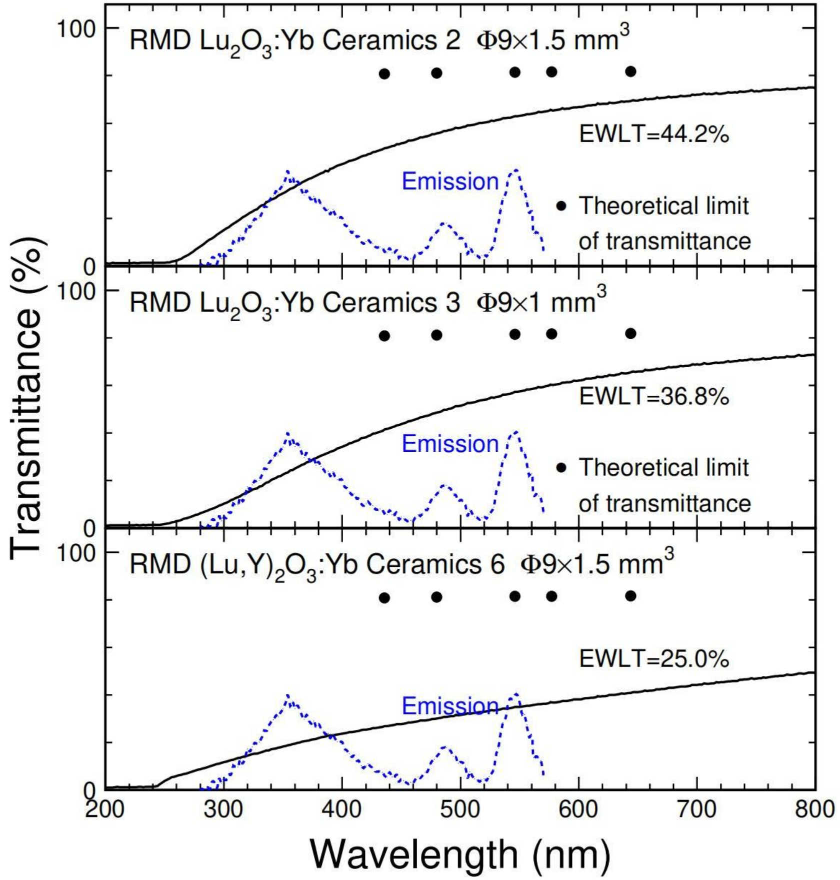
Photo-luminescence and X-ray excited luminescence peaked at ~350 nm and ~550 nm
$(Lu,Y)_2O_3$:Yb sample 6 show poor transmittance, probably due to increased scattering







Light Output/Yield: Lu_2O_3:Yb Ceramics

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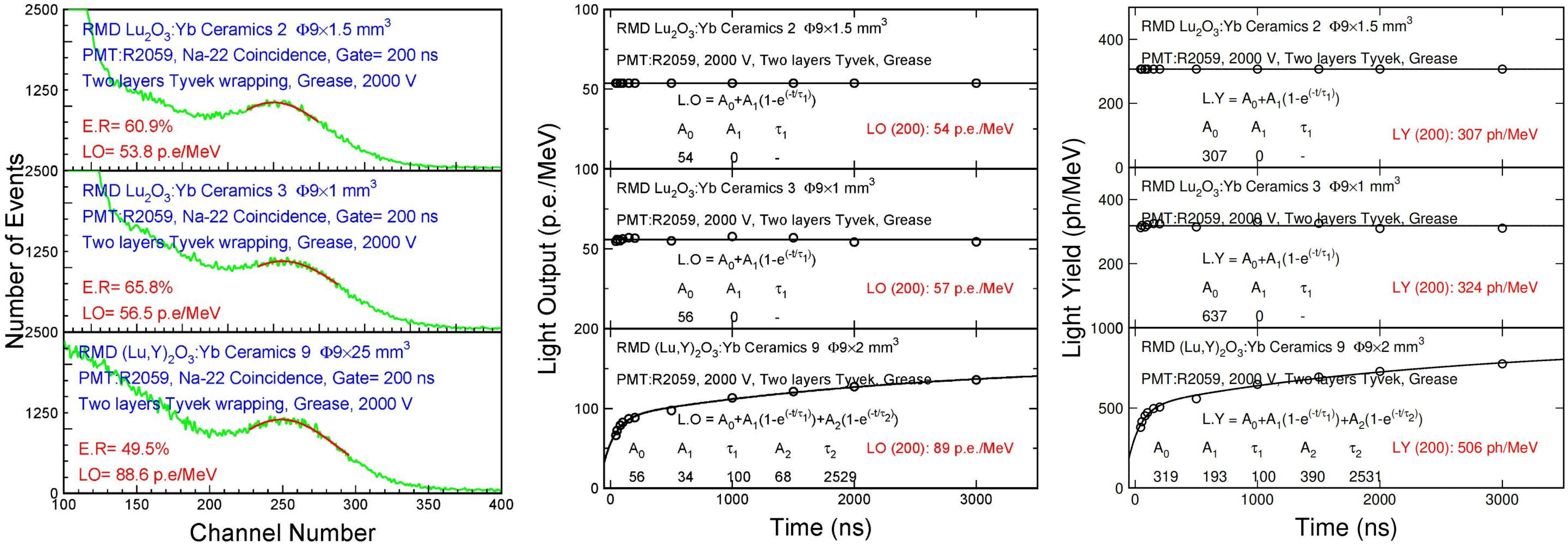
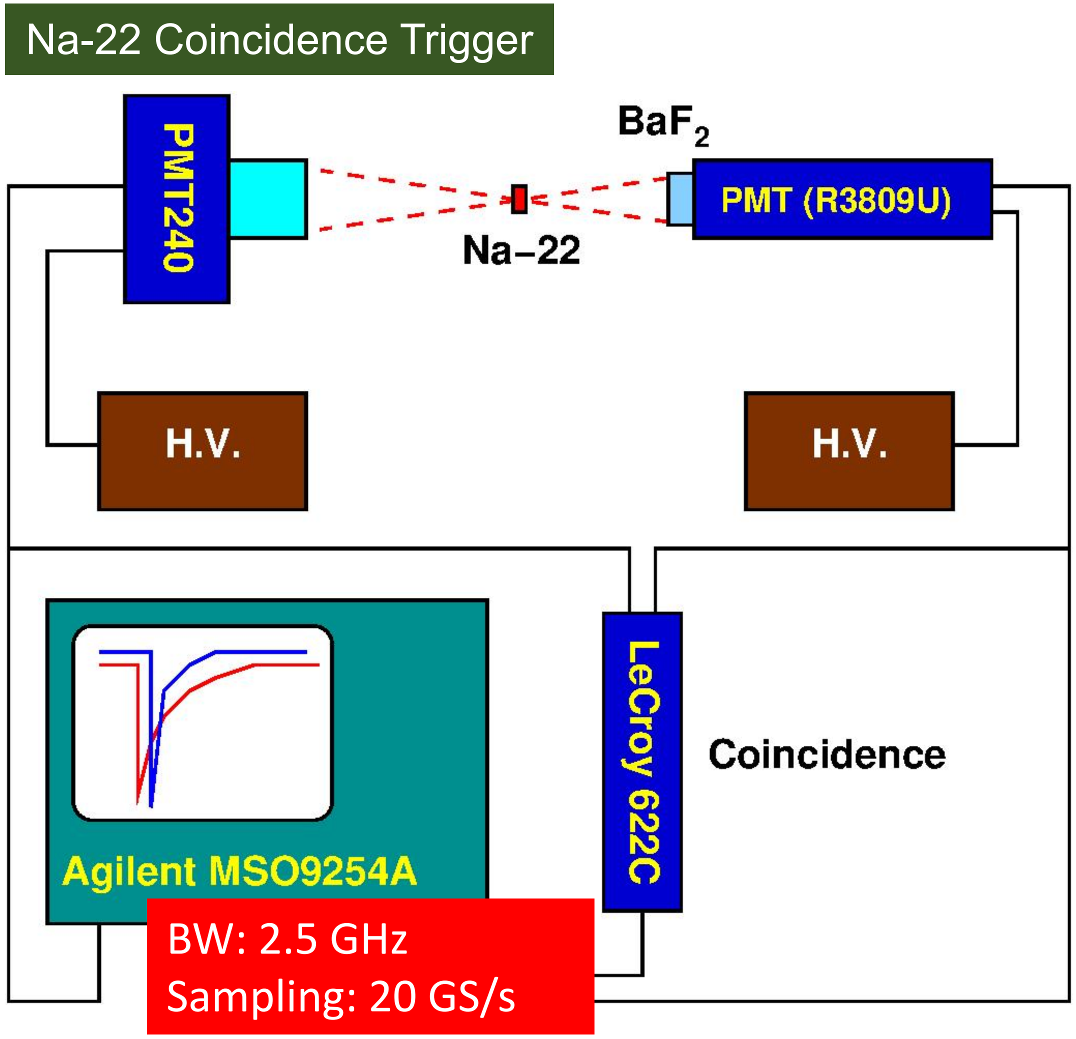
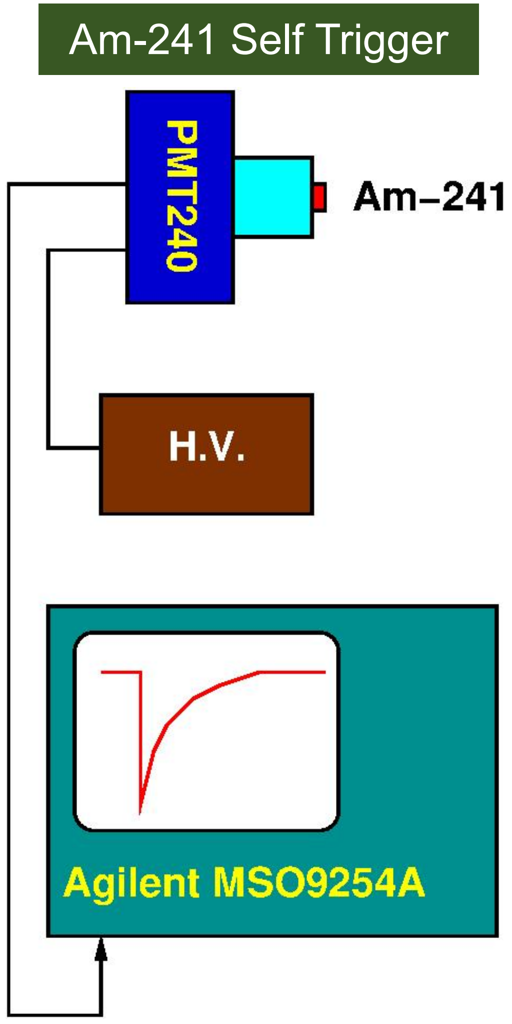
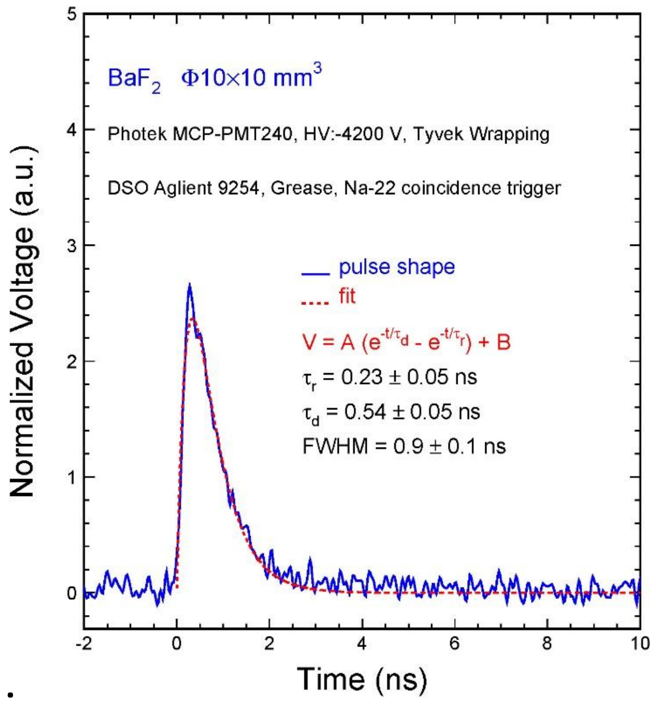
Light yield calculated by taking QE/PDE out of the measured light output
Lu_2O_3:Yb shows light yield up to 320 ph/MeV with negligible slow component
Y admixture increases light output, but introduces slow light of ~100 and ~2,500 ns



An MCP-PMT 240-Based Test Bench







Fitting:

$$V = A(e^{-\frac{t}{\tau_d}} - e^{-\frac{t}{\tau_r}}) + B$$

B: background noise or slow component,

τ_r: rise time,

τ_d: decay time.

Rise, decay and FWHM obtained by fitting temporal response

Decay Time: BaF_2 and Lu_2O_3:Yb

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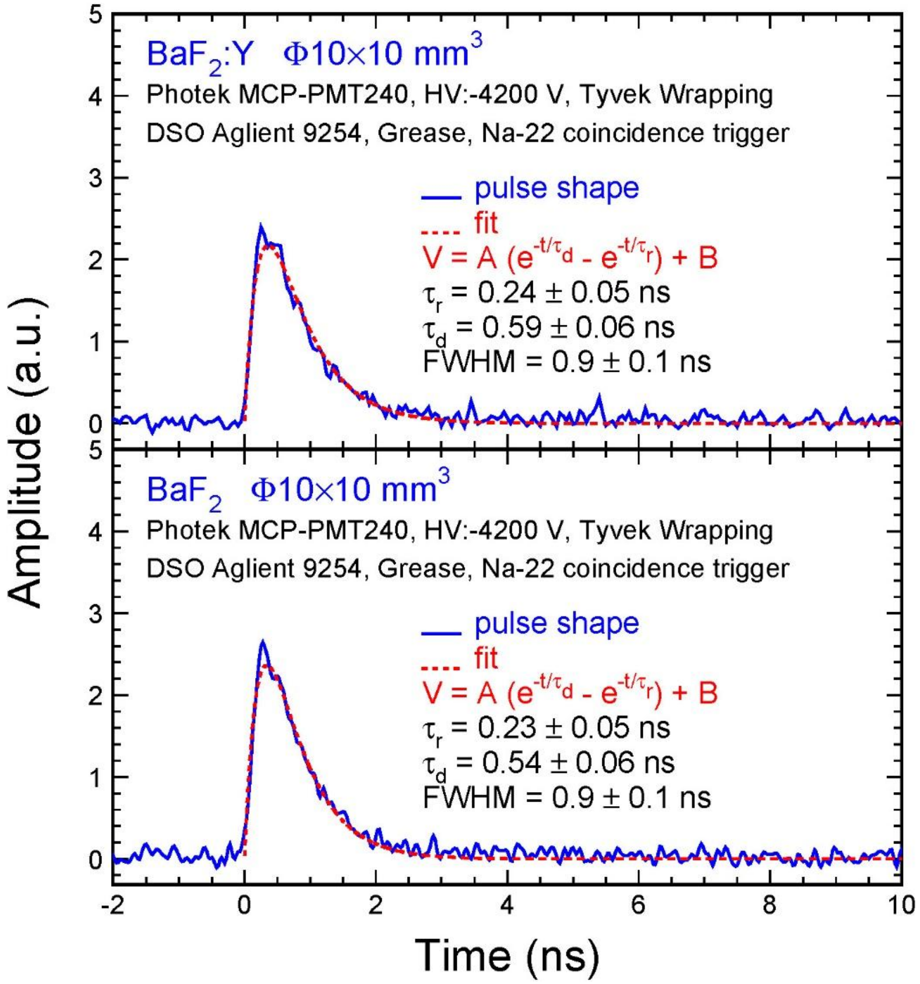
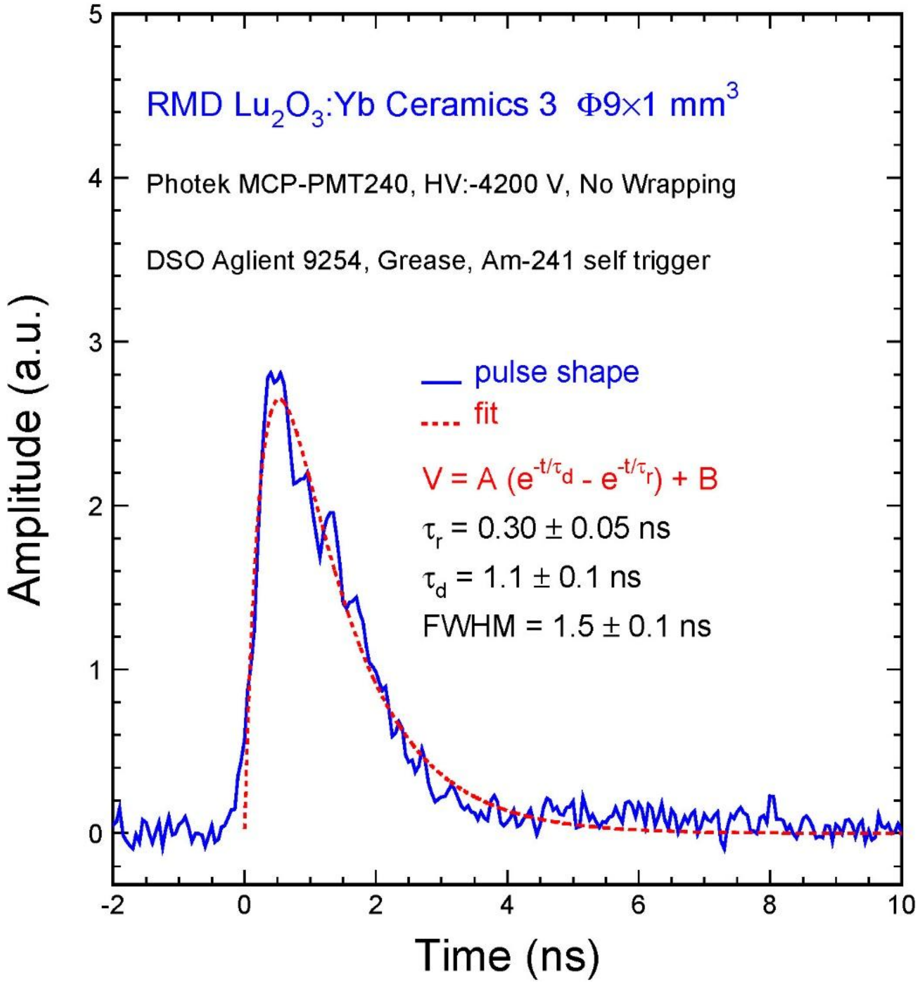
Decay time of 0.5 and 1.1 ns observed for BaF_2 crystals and RMD Lu_2O_3:Yb-3





Radiation Hardness: Lu_2O_3:Yb Ceramics

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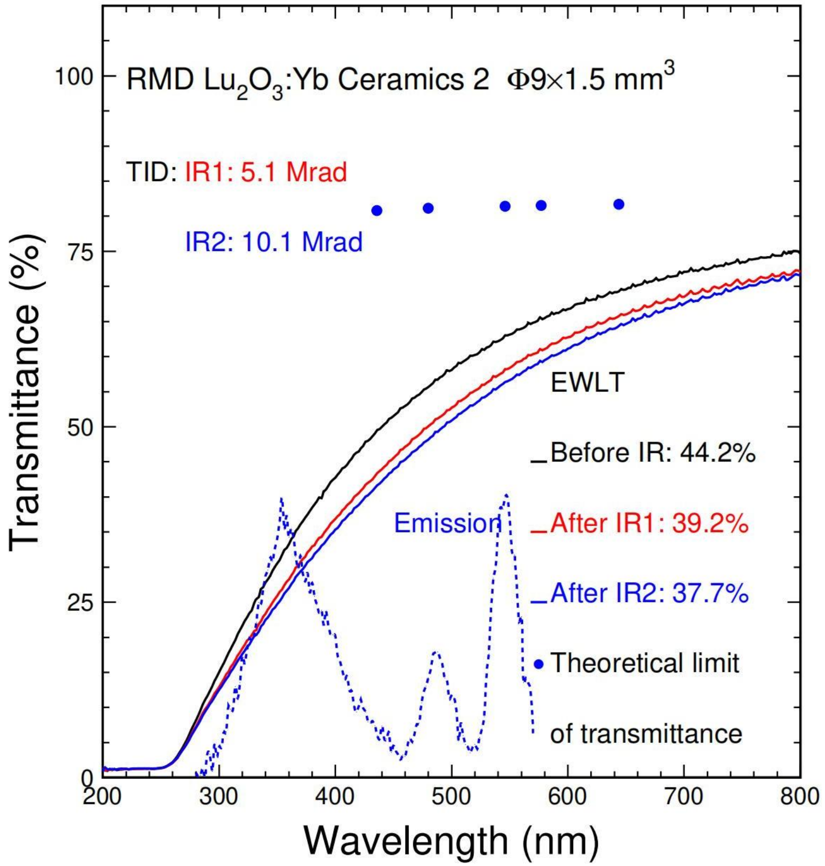
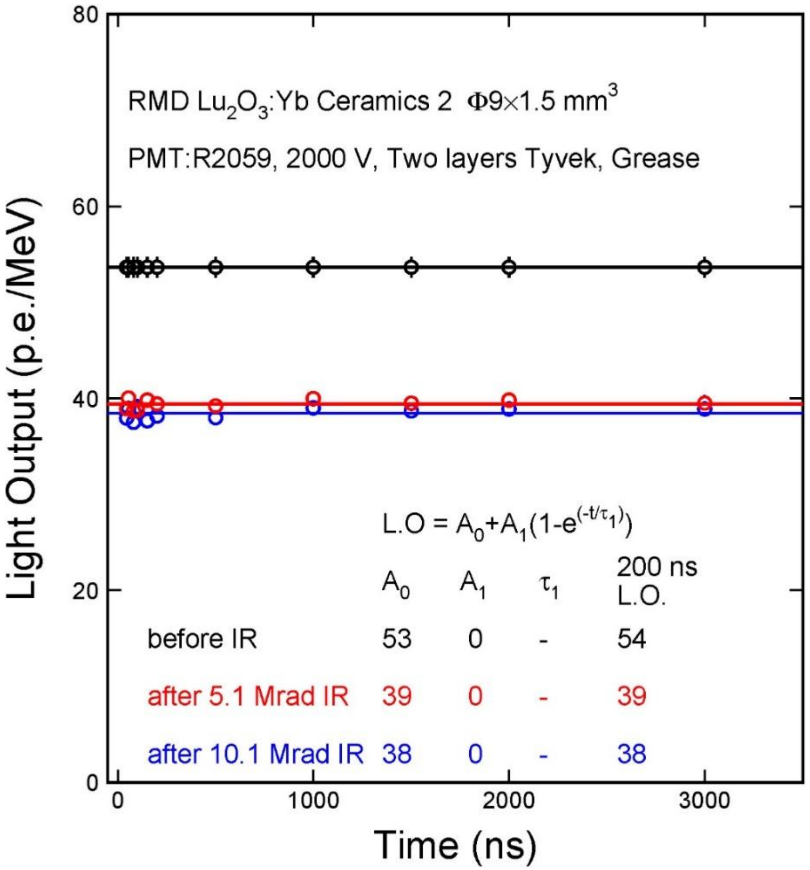
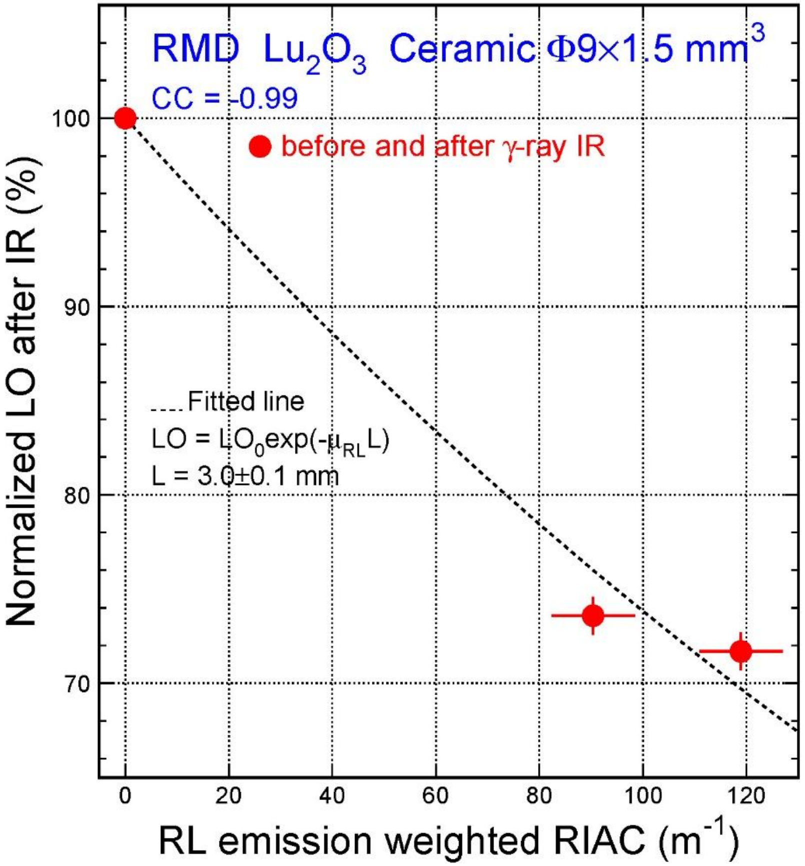
Damage appears saturated after 5.1 Mrad. Dose rate dependence under study
Light output loss is due to induced absorption with a mean light path of 3 mm

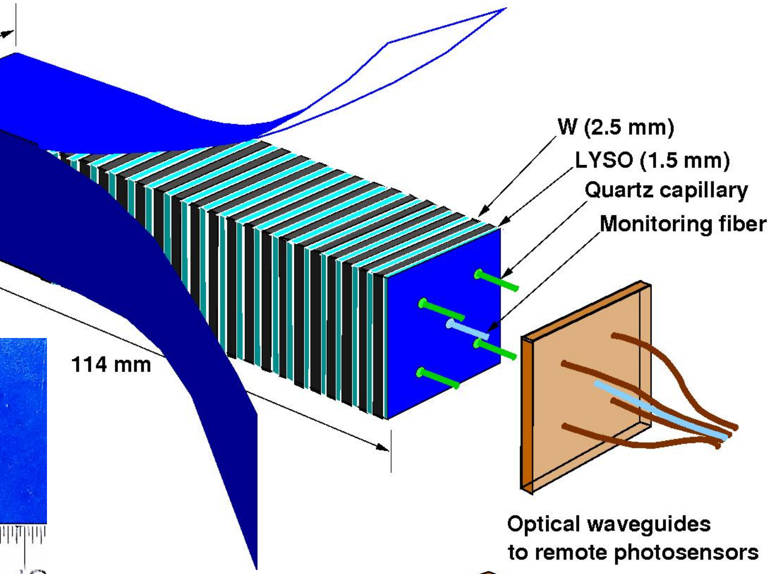


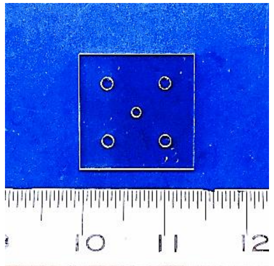


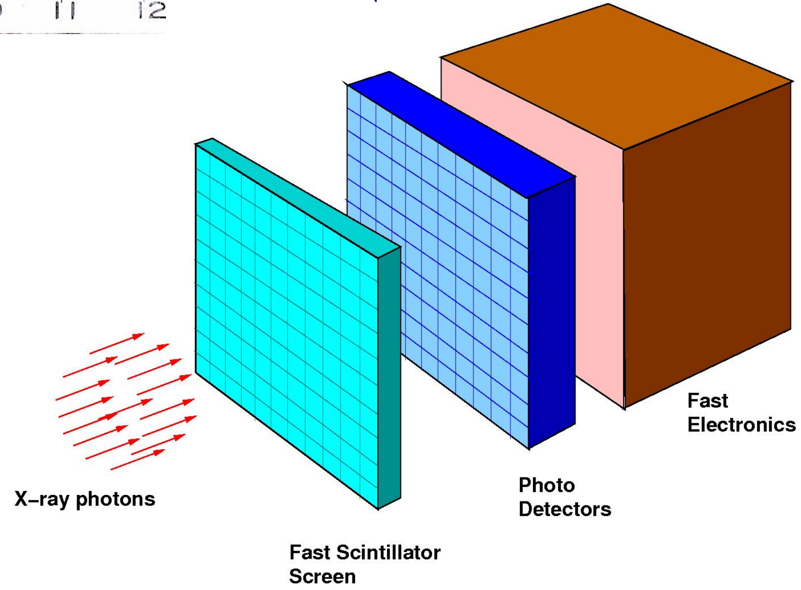


Why LuAG Ceramics?

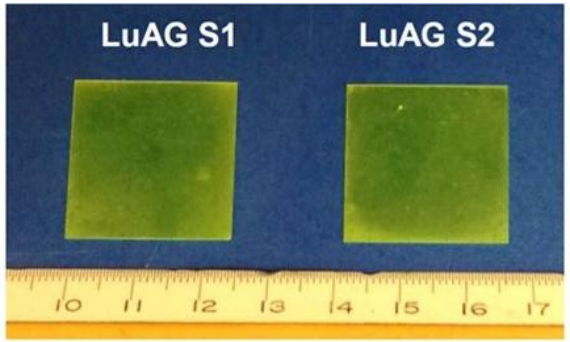
- **Future inorganic scintillators:**
  - **Ultrafast and rad hard scintillators for HL-LHC & FCC-hh;**
  - **Ultrafast scintillators for high rate (Mu2e-II);**
  - **Ultrafast scintillators for GHz hard X-ray imaging in future free electron laser facilities (DMMSC).**
- **Millimeter slices of LYSO:Ce, BaF_2:Y and LuAG:Ce survive the severe radiation environment expected at the HL-LHC with 3,000 fb^{-1}:**
  - **Absorbed dose: up to 100 Mrad,**
  - **Charged hadron fluence: up to 6×10^{14} p/cm^2,**
  - **Fast neutron fluence: up to 3×10^{15} n/cm^2.**
- **LuAG ceramic slices are more cost-effective as compared to crystals :**
  - **Simpler production technology at a lower temperature;**
  - **Higher raw material usage; and**
  - **No need for after growth mechanical processing.**

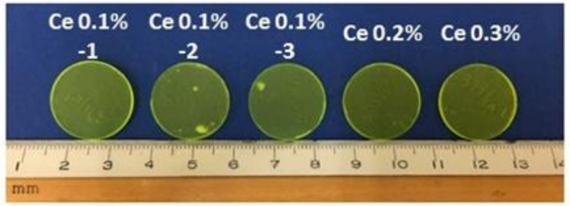


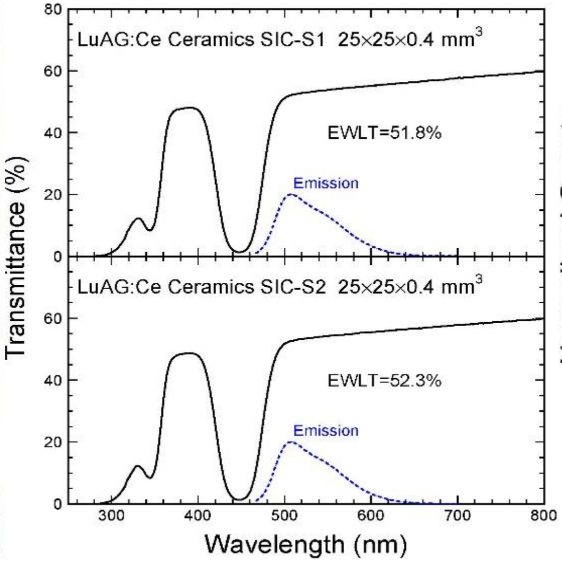


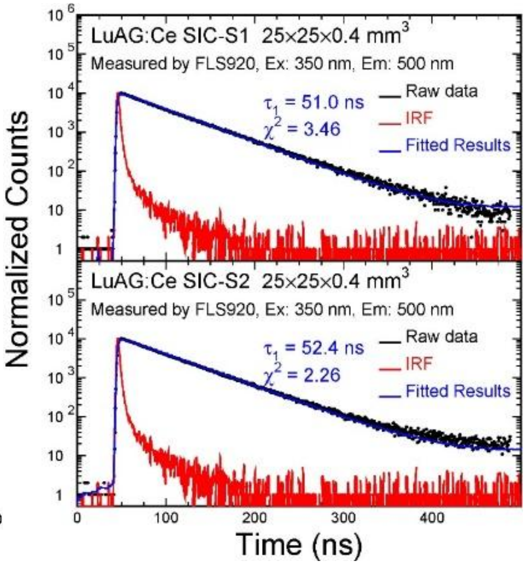


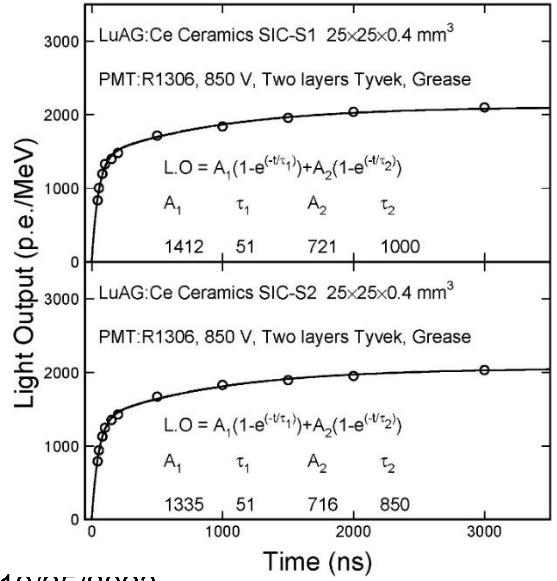
Performance of LuAG:Ce Ceramics

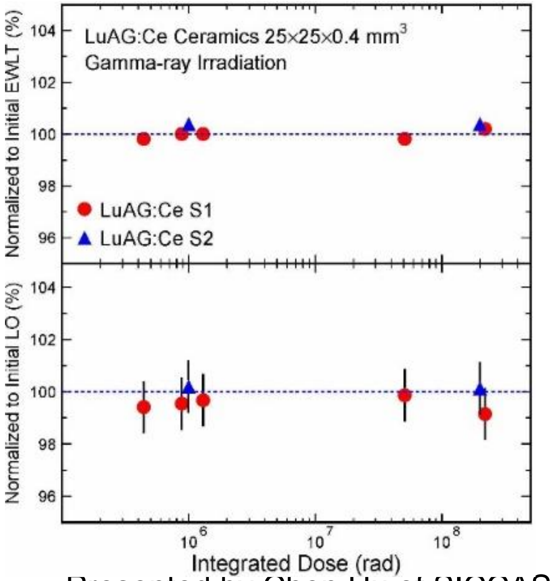


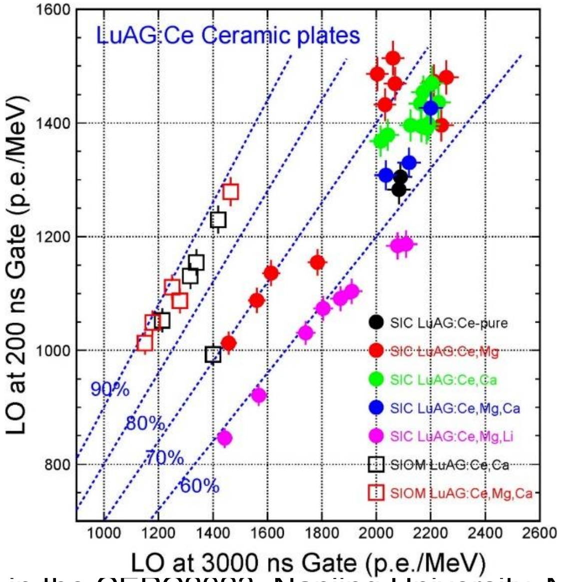












Light output: 1,400 p.e./MeV with a fast decay time of about 50 ns and a slow decay time of about 1 µs. Excellent radiation hardness against ionization dose up to 220 Mrad.

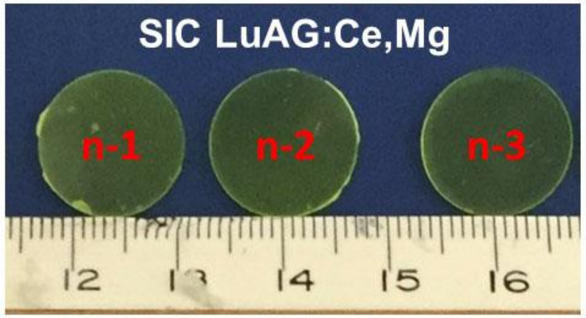
Ca^{2+} co-doping suppresses the slow component. F/T ratio, defined as LO(200 ns) /LO(3 µs), reaches 90%.

C. Hu, et al., *NIMA* 954 (2020) 161723

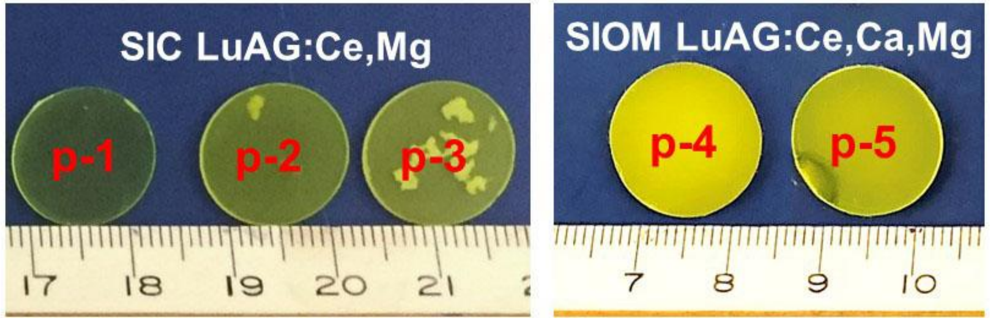
LuAG Samples for Neutron/Proton Irradiation

Mg^{2+} co-doped LuAG:Ce ceramics show a higher light output
Ca^{2+} and Mg^{2+} co-doped LuAG:Ce show a higher F/T ratio

Neutron Irradiation Samples



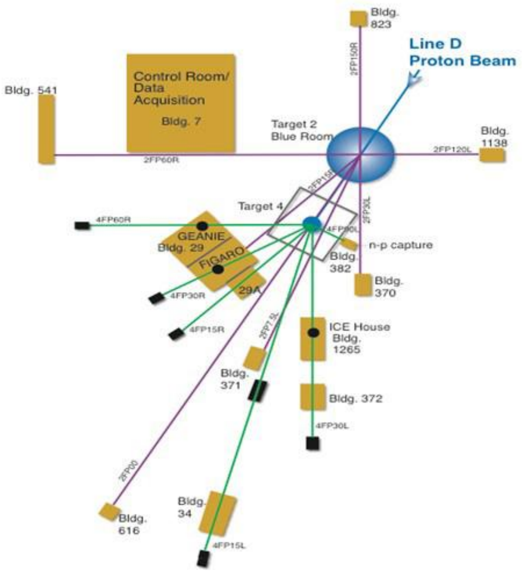
Proton Irradiation Samples

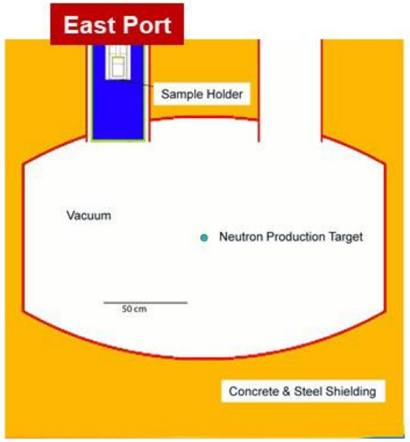


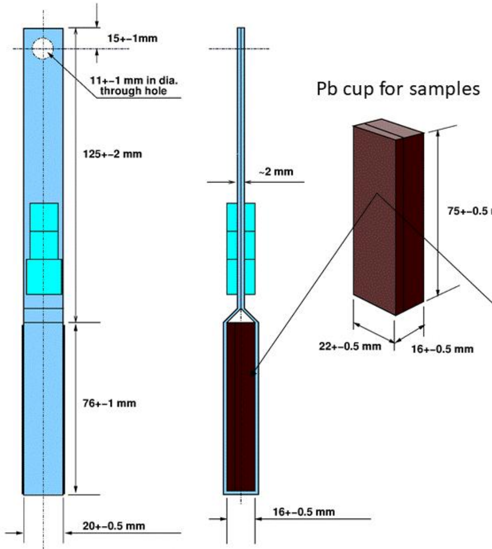
Sample ID	Dimension (mm^3)	200 ns L.O. (p.e./MeV)	F/T ratio (%)	Experiment	Fluence (cm^{-2})
n-1	Φ14.4×1	1474	66.6	LANSCE-7638	1.7×10^{15}
n-2	Φ14.4×1	1479	65.6	LANSCE-7638	3.4×10^{15}
n-3	Φ14.4×1	1514	73.5	LANSCE-7638	6.7×10^{15}

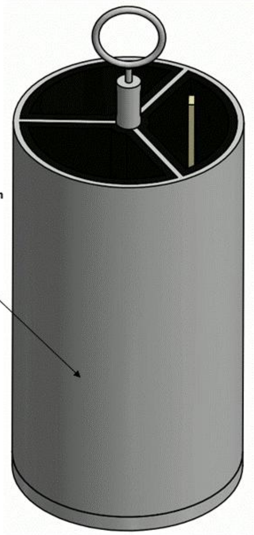
Sample ID	Dimension (mm^3)	200 ns L.O. (p.e./MeV)	F/T ratio (%)	Experiment	Fluence (cm^{-2})
p-1	Φ14.4×1	1486	74.2	CERN	7.1×10^{13}
p-2	Φ14.4×1	1305	62.5	CERN	3.6×10^{14}
p-3	Φ14.4×1	1283	61.6	CERN	1.2×10^{15}
p-4	Φ17×1	1013	88.0	LANSCE-8051	2.4×10^{13}
p-5	Φ17×1	1049	89.0	LANSCE-8051	2.3×10^{14}

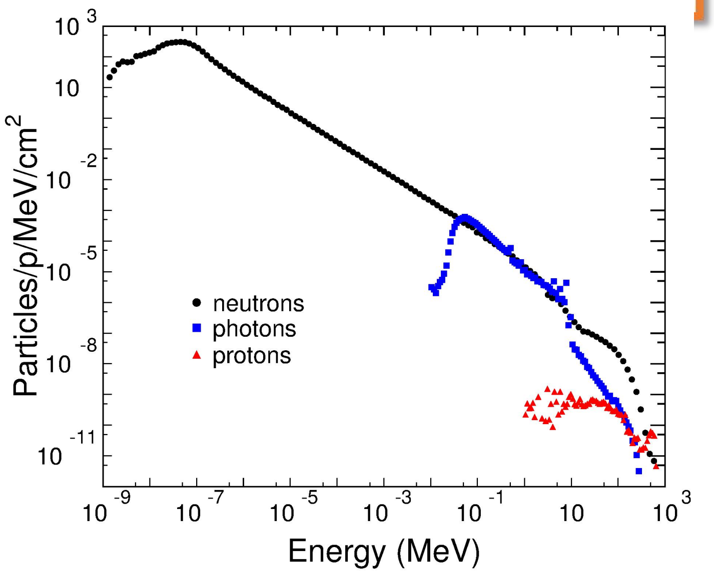
Neutron Irradiation at East Port of LANSCE











1 MeV equivalent neutron fluence is 1.7, 3.4, and 6.7 × 10^{15} cm^{-2} for sample n-1, n-2, and n-3, respectively

Particles	n-1 Fluence (cm^{-2})	n-2 Fluence (cm^{-2})	n-3 Fluence (cm^{-2})
Thermal and Epithermal Neutrons (0 < En < 1 eV)	1.80×10^{15}	3.62×10^{15}	7.14×10^{15}
Slow and Intermediate Neutrons (1 eV < En <1 MeV)	6.57×10^{15}	1.32×10^{16}	2.60×10^{16}
Fast Neutron Fluence (En > 1 MeV)	7.26×10^{14}	1.46×10^{15}	2.88×10^{15}
Very Fast Neutron Fluence (En > 20 MeV)	1.38×10^{14}	2.78×10^{14}	5.49×10^{14}
1 MeV Equivalent Neutron Fluence	**1.69×10^{15}**	**3.40×10^{15}**	**6.71×10^{15}**
Proton Fluence (Ep > 1 MeV)	2.11×10^{12}	4.24×10^{12}	8.38×10^{12}
Photon Dose (rad)	1.05×10^{6}	2.11×10^{6}	4.16×10^{6}

LuAG:Ce after Neutron Irradiations

Small losses in T/LO up to 6.7×10¹⁵ n_{eq}/cm^2 with F/T ratio unchanged

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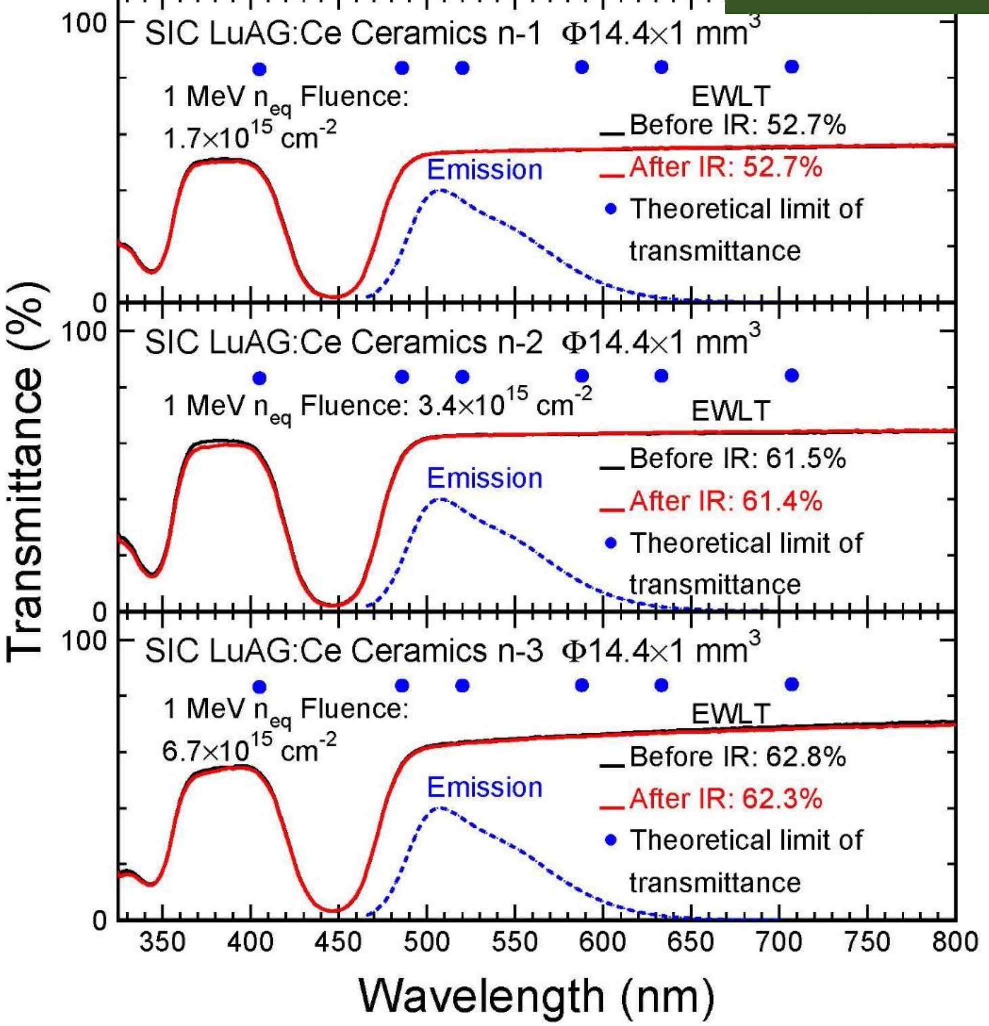

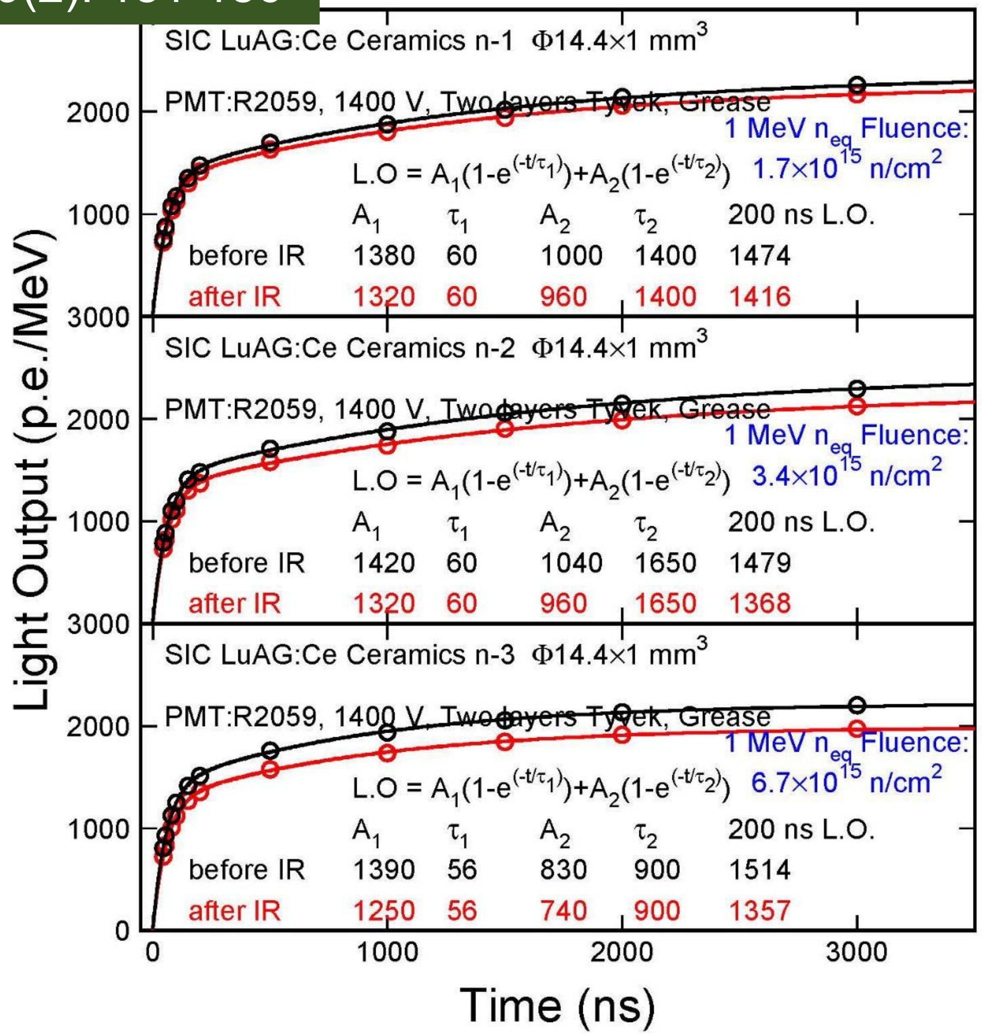

RIAC and LO vs. Neutron Fluence

90% light output remains after an 1 MeV equivalent neutron fluence of 6.7×10^{15} n_{eq}/cm^2
Radiation hardness of LuAG ceramics against neutrons is about a factor of two better than LYSO

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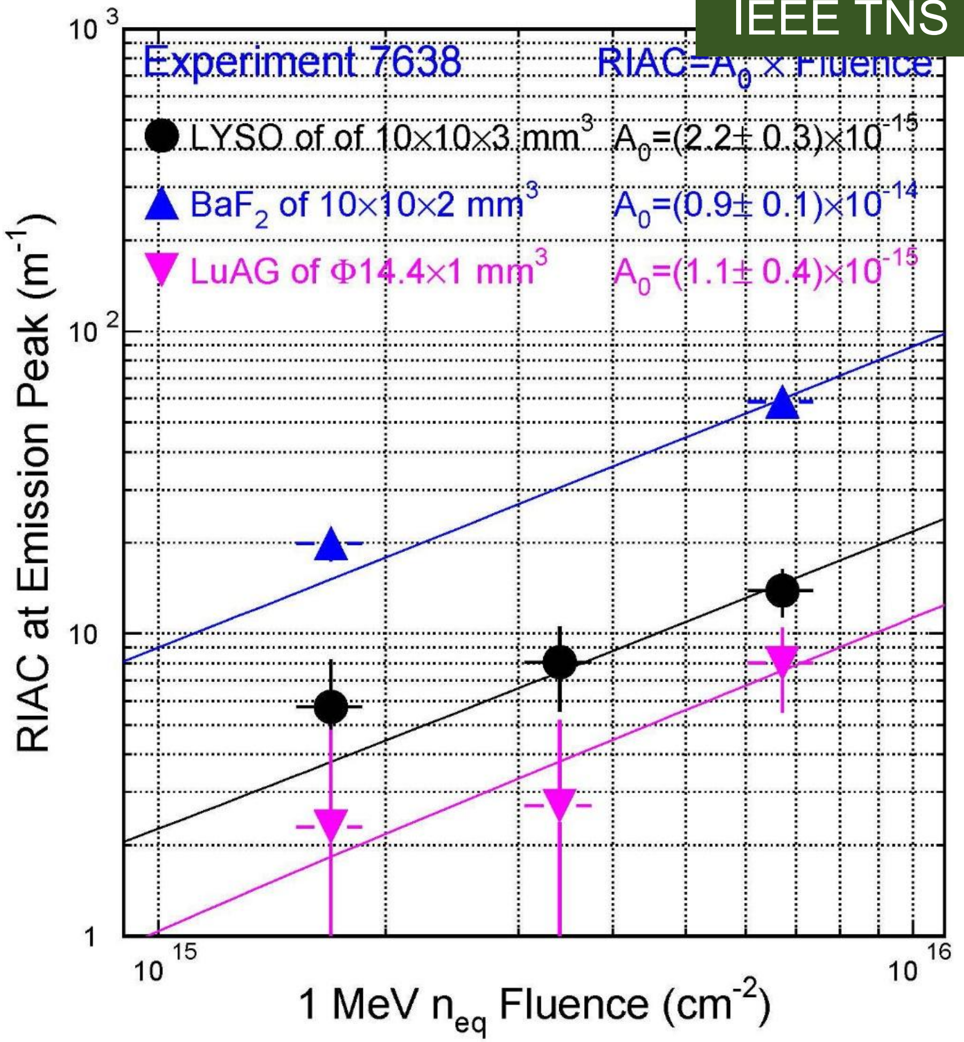
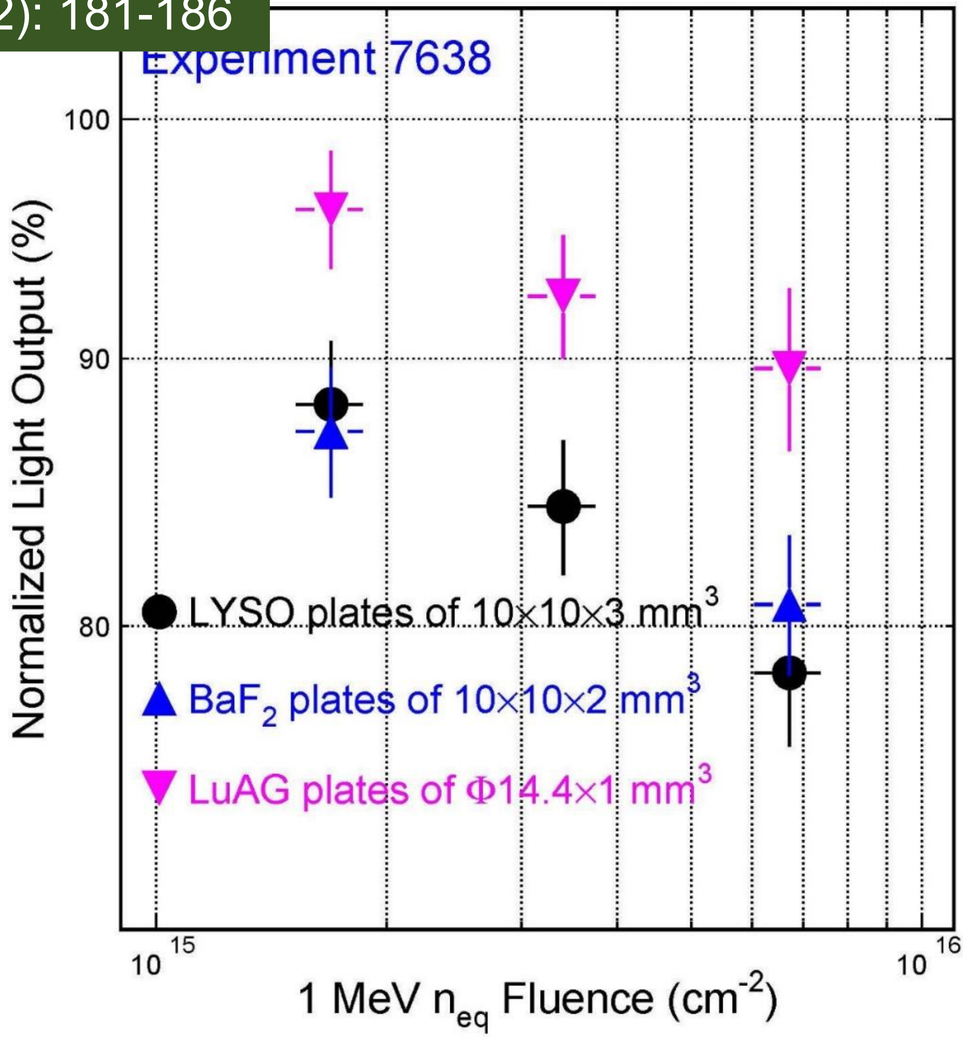
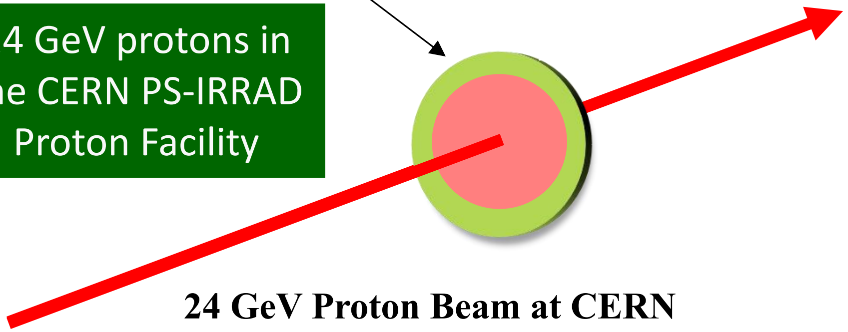




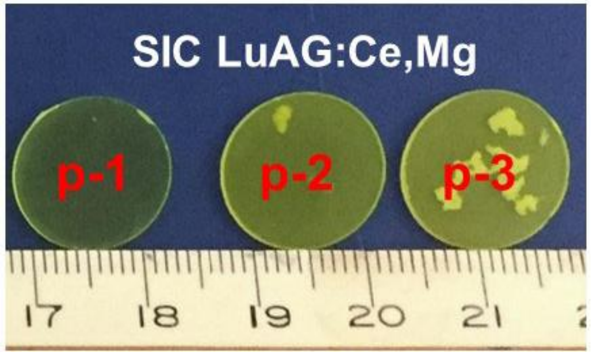
Proton Irradiation at CERN PS & LANSCE

24 GeV protons in the CERN PS-IRRAD Proton Facility

LuAG Plate Φ14.4×1 mm³



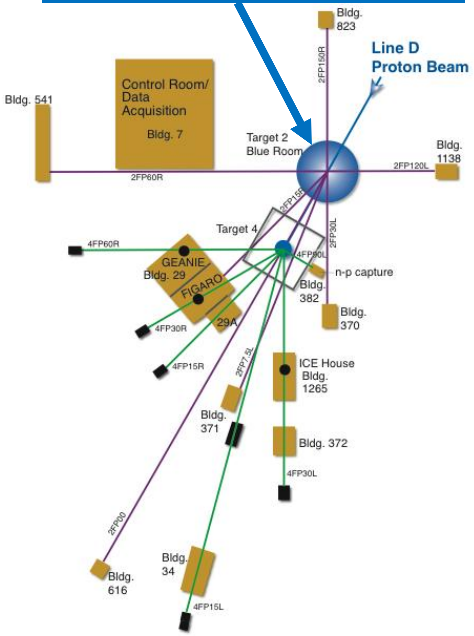
24 GeV Proton Beam at CERN
Gaussian width a FWHM of about 12 mm

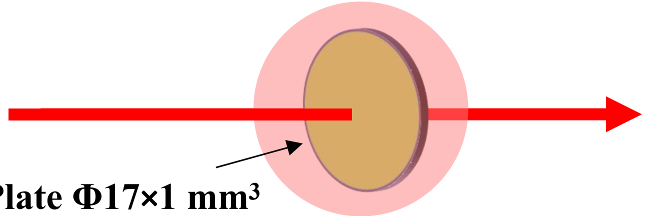


Proton fluence measured by dosimeters of 10×10 and 20×20 mm³ at CERN.

Proton fluence: 7.1×10^{13}, 3.6×10^{14}, and 1.2×10^{15} p cm^{-2} for samples p-1, p-2, and p-3, respectively.

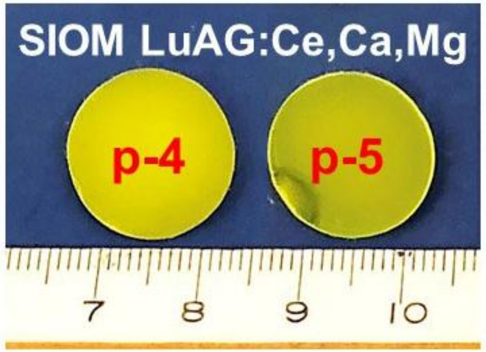
Blue Room in LANSCE





LuAG Plate Φ17×1 mm³

800 MeV proton beam (FWHM= 2.5 cm)



Environment/Source	Proton Flux (p s^{-1} cm^{-2})	Fluence on Crystal (p cm^{-2})
CMS FCAL (η=1.4) at HL-LHC	2.8×10^{5}	2.5×10^{13} / 3000 fb^{-1}
CMS FCAL (η=3.0) at HL-LHC	2.3×10^{6}	2.1×10^{14} / 3000 fb^{-1}
WNR facility of LANSCE	**Up to 2×10^{10}**	**Up to 3×10^{15}**

Proton Fluence: 2.4×10^{13} and 2.3×10^{14} p/cm² applied to samples p-4 and p-5, respectively

LuAG:Ce after Proton Irradiations

Small loses in T/LO after 1.2×10^15 p/cm^2 by 24 GeV protons at CERN and after 2.3×10^14 p/cm^2 by 800 MeV protons at LANSCE with F/T unchanged

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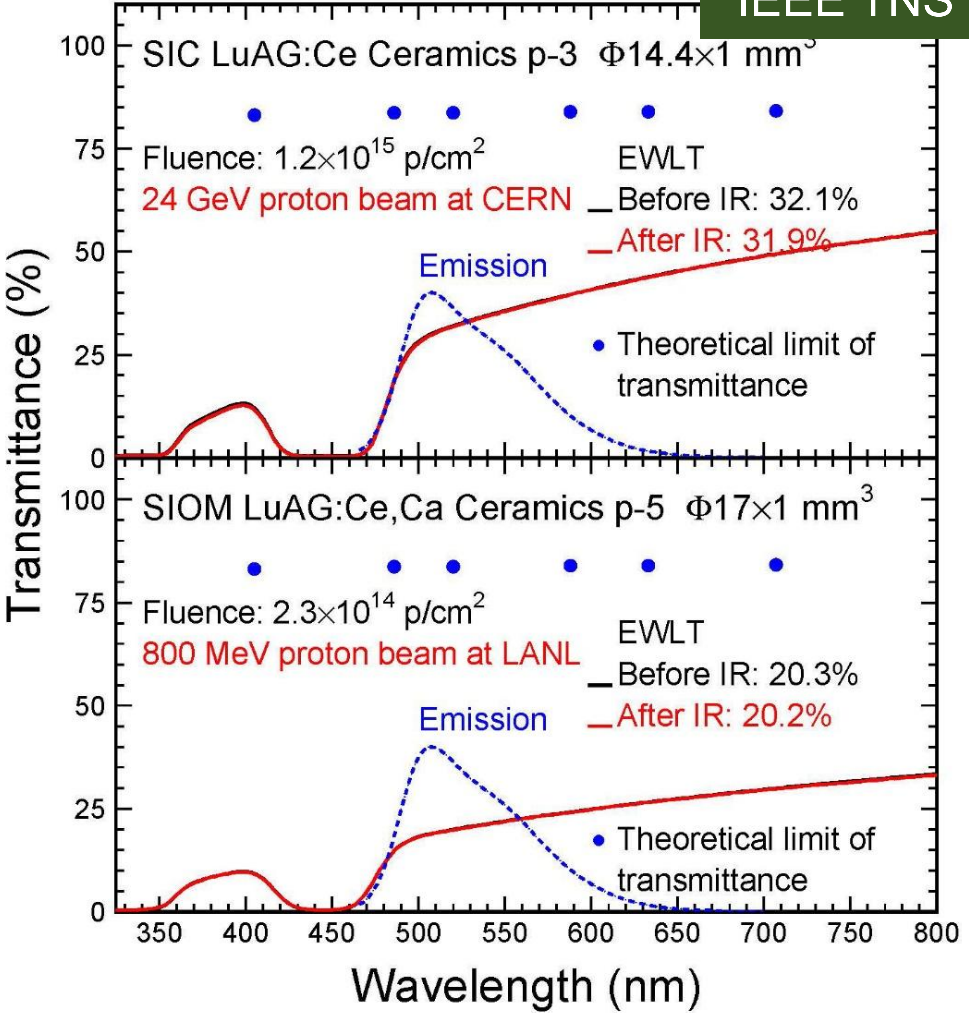

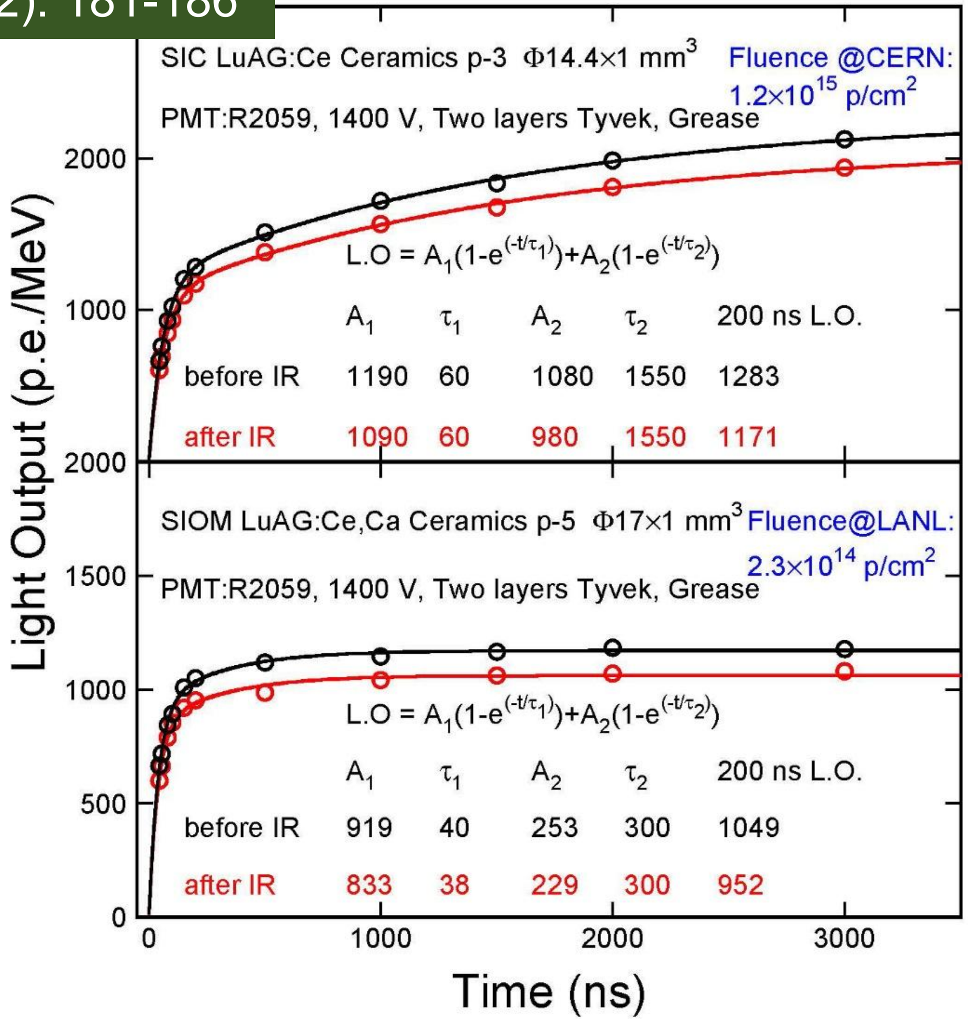

RIAC and LO vs. Proton Fluence

Radiation hardness of LuAG ceramics against protons is also a factor of two better than LYSO
90% light output remains after a proton irradiation fluence up to 1.2×10^{15} p/cm^2

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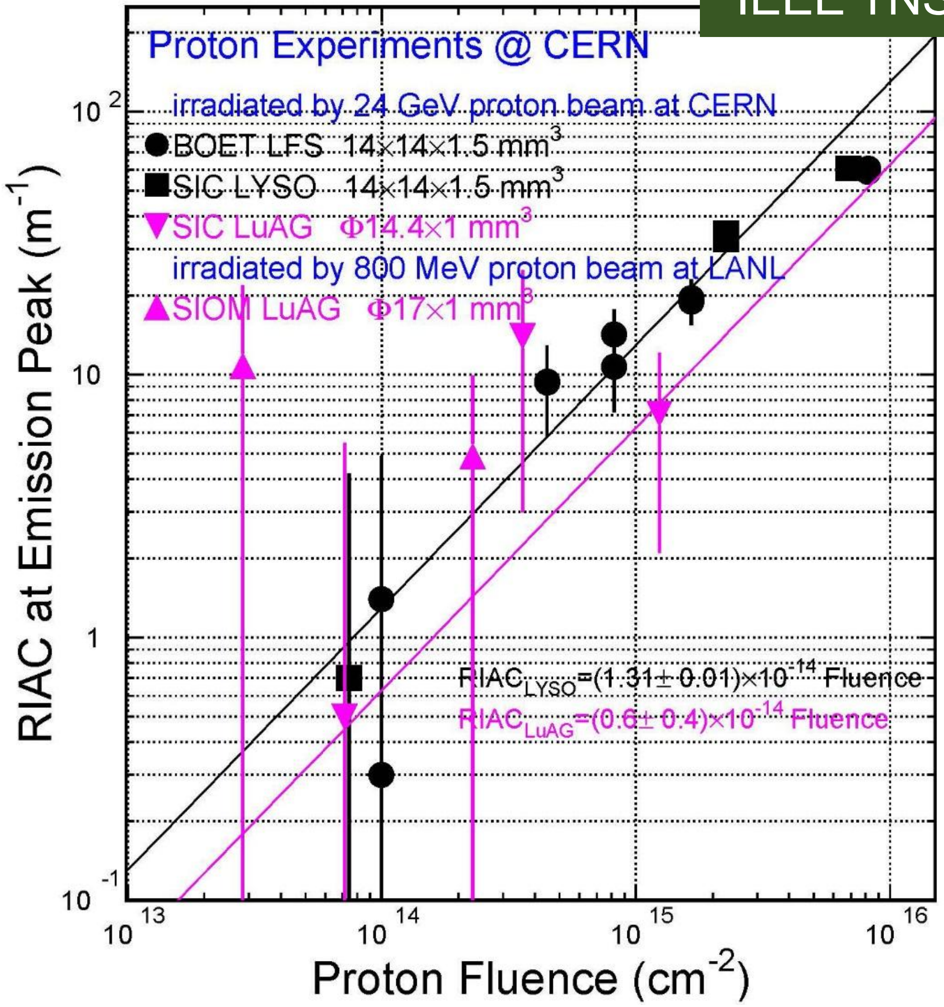
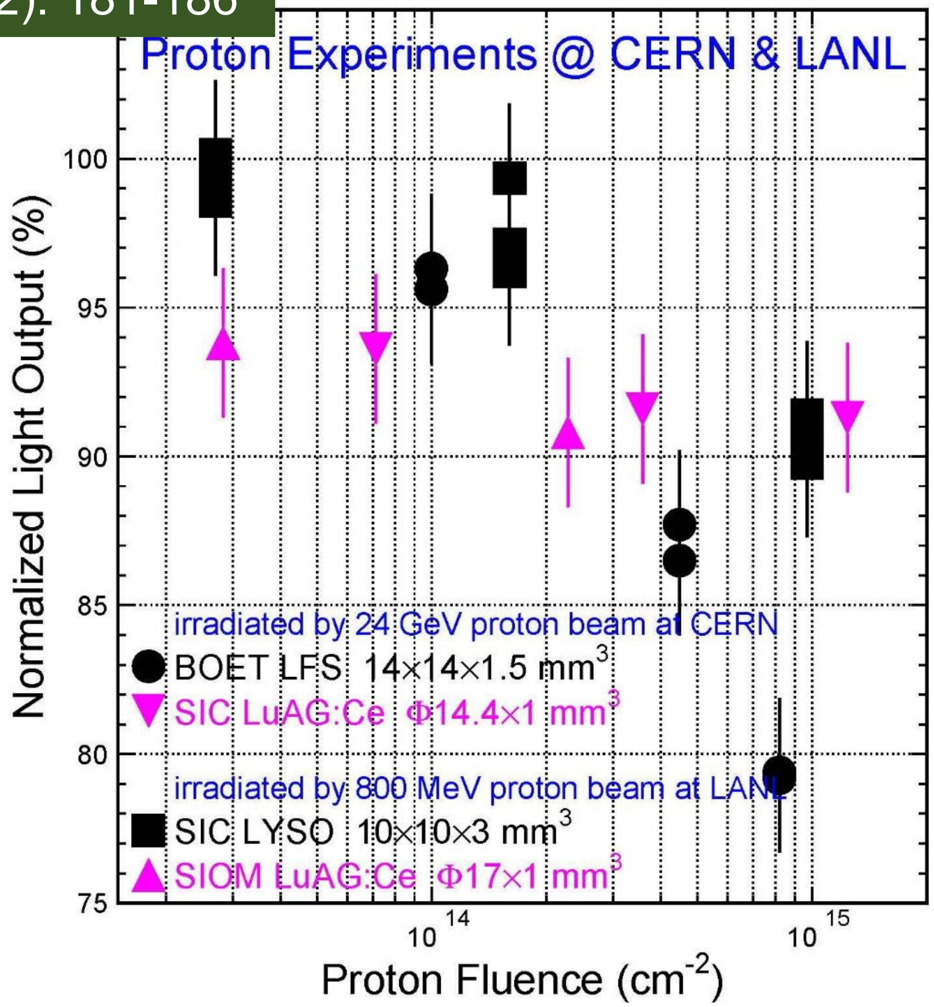
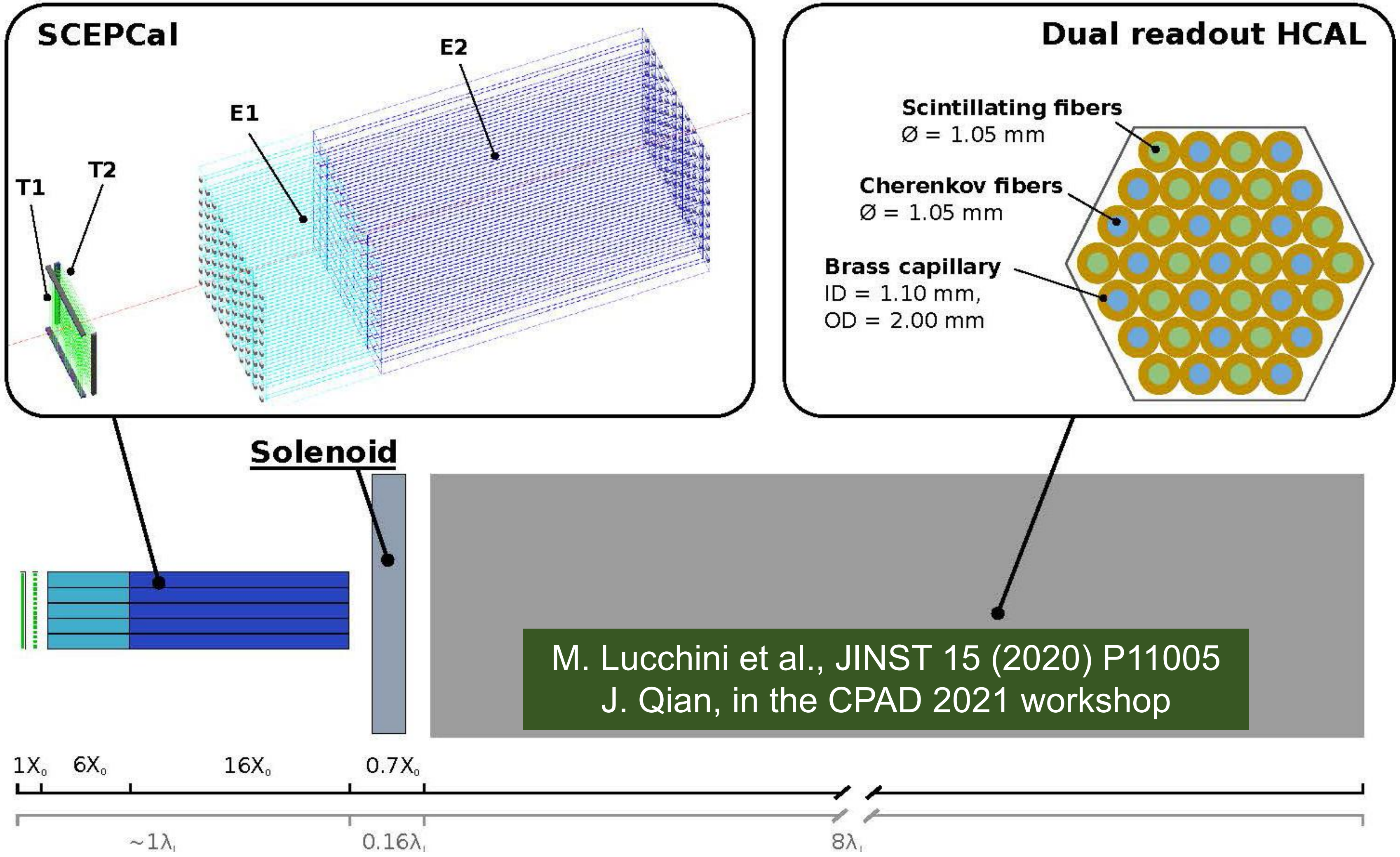




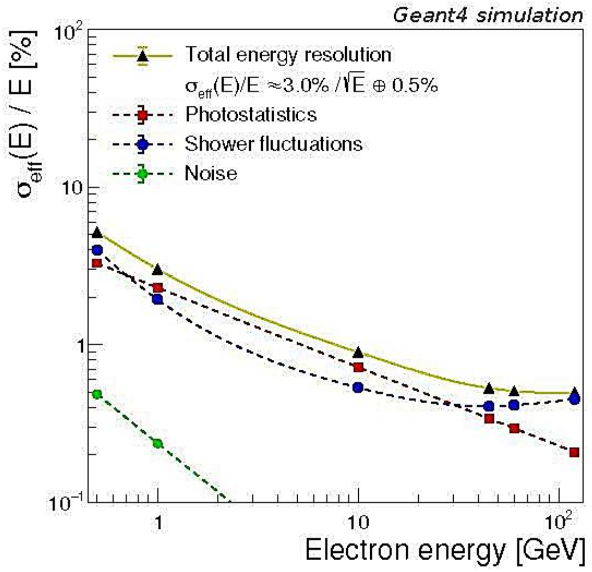
Calvision: A Longitudinally Segmented Crystal ECAL

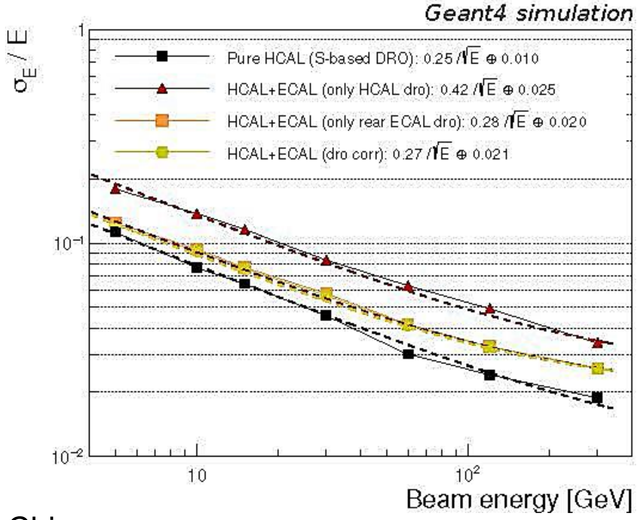
Since 1928 SICCAS

Aiming at excellent EM and jet resolutions for Higgs Factory

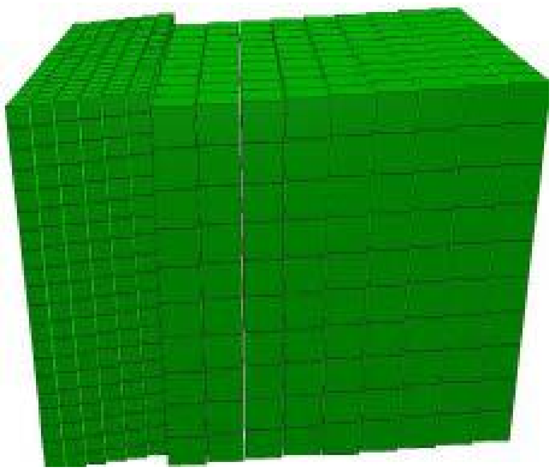


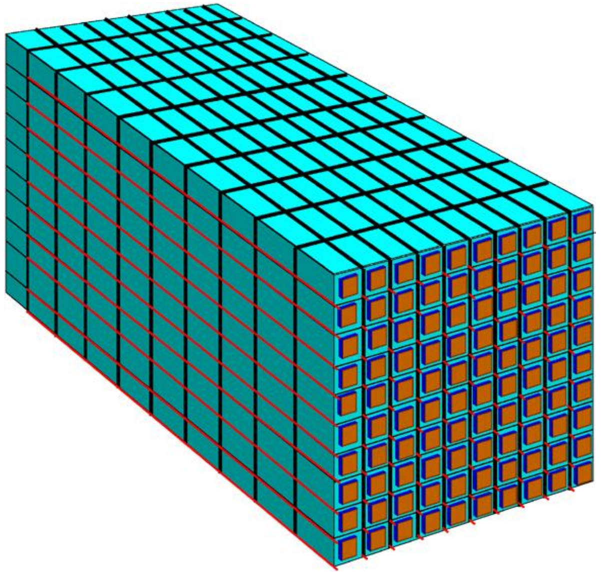
M. Lucchini et al., JINST 15 (2020) P11005
J. Qian, in the CPAD 2021 workshop





HHCAL : A Total Absorption Hadron Calorimeter

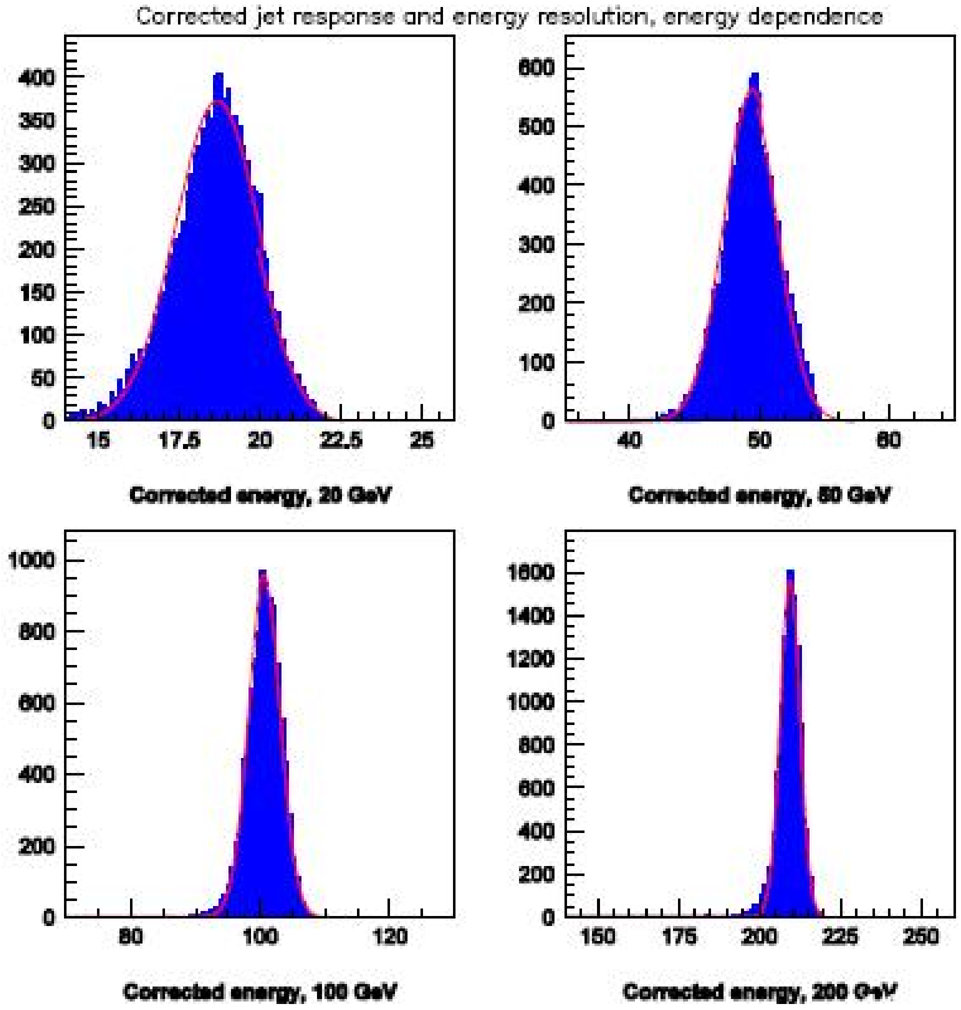




A. Para, H. Wenzel, and S. McGill in Callor2012 Proceedings;
A. Benaglia *et al.*, IEEE TNS **63** (2016) 574:
Jet energy resolution of 20%/√E is achievable by HHCAL with dual readout of S/C or dual gate

See presentations by H. Wenzel and M. Demarteau in this session

Can we afford?



Inorganic Scintillators for HHCAL

	BGO	BSO	PWO	PbF_2	PbFCl	Al_2O_3:Ti	AFO glass	DSB:Ce glass[1]	BGS glass[2]	GS glass[3]	DSB:Ce,Gd glass[4,5]	HFG glass[6]
Density (g/cm^3)	7.13	6.8	8.3	7.77	7.11	3.98	4.6	3.8	4.2	6.03	4.7 - 5.4[d]	5.95
Melting point (℃)	1050	1030	1123	824	608	2040	980[7]	1420[8]	1550	?	1420[8]	570
X_0 (cm)	1.12	1.15	0.89	0.94	1.05	7.02	2.96	3.36	2.61	1.51	2.14	1.74
R_M (cm)	2.23	2.33	2.00	2.18	2.33	2.88	2.89	3.52	3.33	2.51	2.56	2.45
λ_I (cm)	22.7	23.4	20.7	22.4	24.3	24.2	26.4	32.8	31.9	24.9	24.2	23.2
Z_{eff} value	72.9	75.3	74.5	77.4	75.8	11.2	42.8	44.4	51.2	57.6	48.7	56.9
dE/dX (MeV/cm)	8.99	8.59	10.1	9.42	8.68	6.75	6.84	5.56	5.90	7.95	7.68	8.24
Emission Peak[a] (nm)	480	470	425 420	\	420	300 750	365	440	430	396	440 460	325
Refractive Index[b]	2.15	2.68	2.20	1.82	2.15	1.76	\	\	\	?	\	1.50
Relative Light Output by PMT[a,c]	7,500	1,500	130	\	150	7,900	450	~500	2,500	800	1,300	150
LY (ph/MeV)[d]	300	100	30 10	\	3	300 3200	40	180 30	400 90	1200 260	120, 400 50	25 8
Decay Time[a] (ns)	-0.9	?	-2.5	\	?	?	?	-0.04	0.3	?	?	-0.37
d(LY)/dT (%/°C)[d]	6.0	7.0	7.5	6.0	?	0.6	?	2.0	2.0	?	2.0	high cost

a. Top line: slow component, bottom line: fast component.
b. At the wavelength of the emission maximum.
c. Relative light yield normalized to the light yield of BGO
d. At room temperature (20°C) with PMT QE taken out.

arXiv:2203.06731

Low density crystals/glasses

Summary

- Future HEP experiments at the energy and intensity frontiers present stringent challenges to inorganic scintillators in radiation hardness, ultrafast time response and cost.
- Inorganic scintillators in ceramic form have attracted a broad interest due to its lower fabrication temperature, effective usage of raw material, and no need for aftergrowth mechanical processing.
- Lu_2O_3:Yb transparent ceramics show PL and XEL emission peaked at ~350 and ~550 nm.
- Lu_2O_3:Yb ceramics show light yield up to 320 ph/MeV with negligible slow component. Mixing Lu_2O_3 with Y_2O_3 appears increase light yield in 200 ns to 500 ph/MeV with significant slow component of 100 and 2,500 ns decay time.
- Sub-nanosecond decay time of 1.1 ns was observed by using MCP-PMT.
- Mg^{2+} co-doping in LuAG ceramics improves light output, while Ca^{2+} and Mg^{2+} co-doping improves F/T ratio.
- LuAG ceramics were found to have a factor of two better radiation hardness than LYSO crystals against both neutrons and protons. With 90% of the light output remains in 1 mm thick samples after neutron and proton irradiation up to 6.7×10^{15} n_{eq}/cm^2 and 1.2×10^{15} p/cm^2 respectively it is promising for applications at the HL-LHC and FCC-hh.
- R&D is needed for cost-effective mass produced inorganic scintillators such as scintillating glass.

Acknowledgements: DOE HEP Award DE-SC0011925 and SBIR Award DE-SC0021686