Neutron and Proton-Induced Radiation Damage in LuAG Scintillating Ceramics

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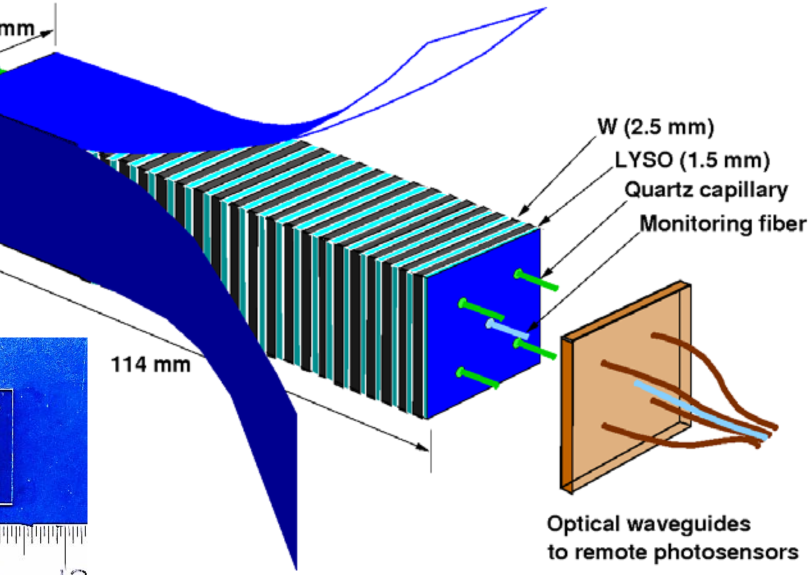
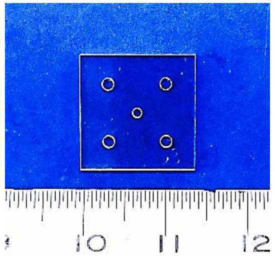
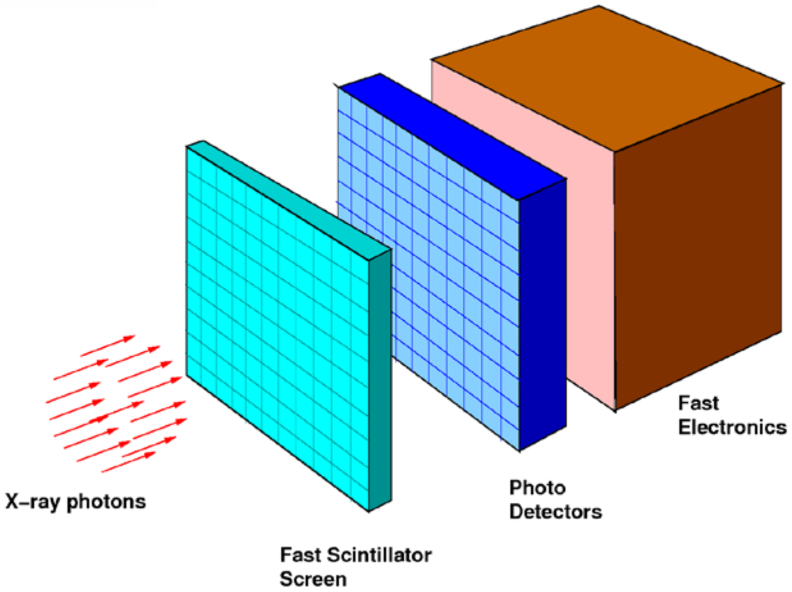
Why LuAG Ceramics?

- **Future applications of 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_{eq}/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.**







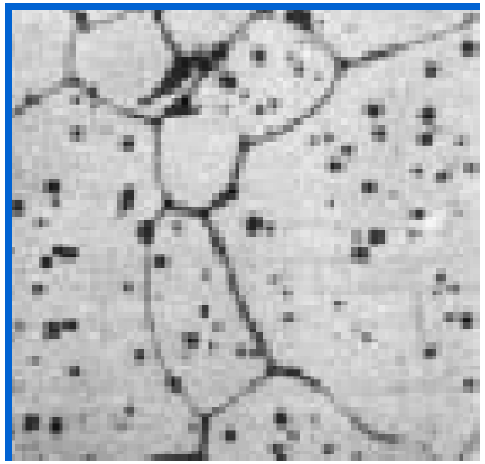
Transparent Ceramics with Cubic Structure

Ceramic Scintillators	Crystal system	Transparency	Density (g/cm^3)
$Y_{1.34}Gd_{0.6}Eu_{0.06}O_3$	Cubic	Transparent	5.92
Gd_2O_2S:Pr,Ce,F	Hexagonal	Translucent	7.34
$Gd_3Ga_5O_{12}$:Cr,Ce	Cubic	Transparent	7.09
$BaHfO_3$:Ce	Cubic	Opaque	8.35

Annu. Rev. Mater. Sci. 1997. 27:69–88

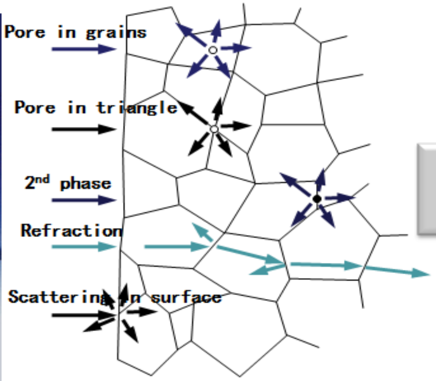
Cost-effective transparent ceramics are a strong competitor to crystals

Raw Materials: >99.99% purity
↓ α-Al_2O_3, Y_2O_3, CeO_2 , Pr_6O_{11}, commercial powders
Sintering aids (TEOS/ MgO) Solvent: Ethyl Alcohol
Ball milling:
↓ 12 hours
Dry and Sieving: Powder
↓ 80ºC&200Mesh
Dry Pressing: Green body
↓
Cold Isostatic Pressing: 250MPa
→
Vacuum Sintering: Densification
↓ ~1550ºC~1770ºC
Annealing
↓ ~1450ºC, 20h
Optical Polishing
↓
Transparent Ceramics

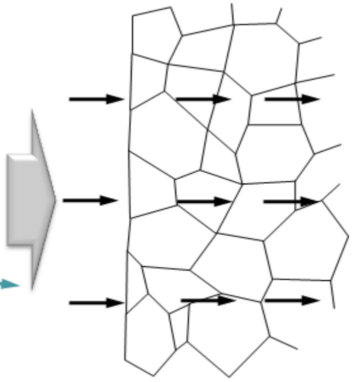


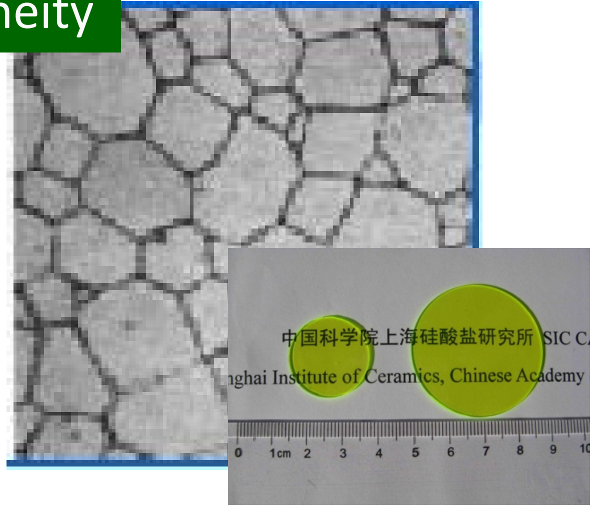
Incident light →

Anisotropy



Homogeneity





Opaque or translucent

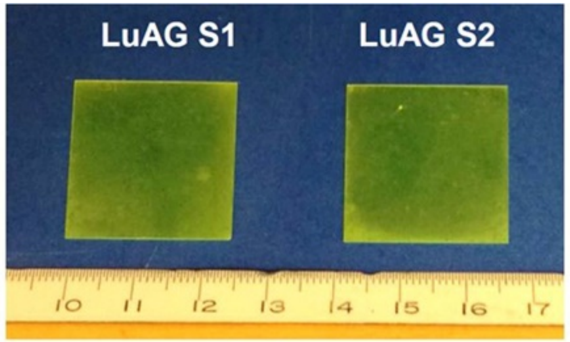
Transparent

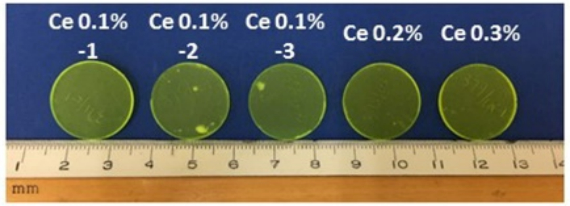
Fast Inorganic Scintillators

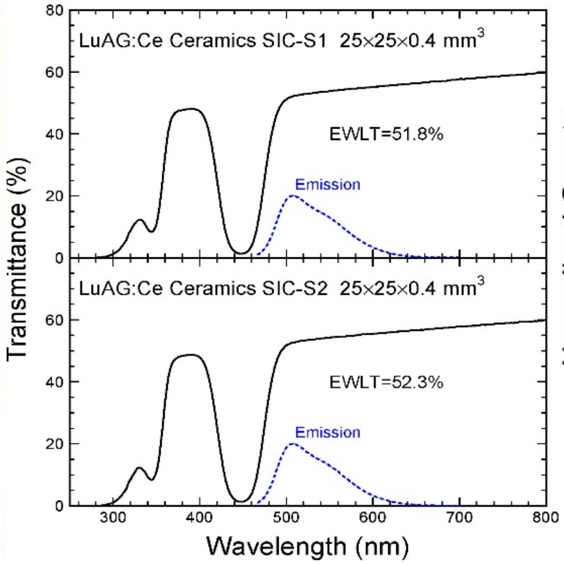
	LYSO:Ce	LSO:Ce,Ca[a]	LuAG:Ce[b]	LuAG:Pr[c]	GGAG:Ce[d]	GLuGAG:Ce[g]	GYGAG:Ce[h]	$SrHfO_3$:Ce[i]	$BaHfO_3$:Ce[i]	$CeBr_3$[k]	$LaBr_3$:Ce[k]
Density (g/cm^3)	7.4	7.4	6.76	6.76	6.5	6.80	5.80	7.56	8.5	5.23	5.29
Melting points (°C)	2050	2050	2060	2060	1850[e]	>1900[e]	1850[e]	2730	2620	722	783
X_0 (cm)	1.14	1.14	1.45	1.45	1.63	1.38	2.11	1.17	0.98	1.96	1.88
R_M (cm)	2.07	2.07	2.15	2.15	2.20	1.57	2.43	2.03	1.87	2.97	2.85
λ_I (cm)	20.9	20.9	20.6	20.6	21.5	20.8	22.4	20.6	19.4	31.5	30.4
Z_{eff}	64.8	64.8	60.3	60.3	51.8	55.2	45.4	60.9	62.9	45.6	45.6
dE/dX	9.55	9.55	9.22	9.22	8.96	9.28	8.32	9.80	10.7	6.65	6.90
λ_{peak} (nm)	420	420	520	310	540	550	560	410	400	371	356
Refractive Index	1.82	1.82	1.84	1.84	1.92[f]	1.92[f]	1.92[f]	2.0	2.1	1.9	1.9
Normalized Light Yield	100	116	83	73	115	161	167	133	133	99	153
Total Light yield (ph/MeV)[a]	30,000	34,800	25,000	22,000	34400	48,200	50,000	5,000[j]	5,000[j]	30,000	46,000
Decay time (ns)[a]	40	31	46	20	53	84 148	100	42	25	17	20
Light Yield in 1st ns (photons/MeV)	740	950	540	1,100	640	570	500	120	200	1,700	2,200
Issues					high thermal neutron x-section			incongruent		hygroscopic	

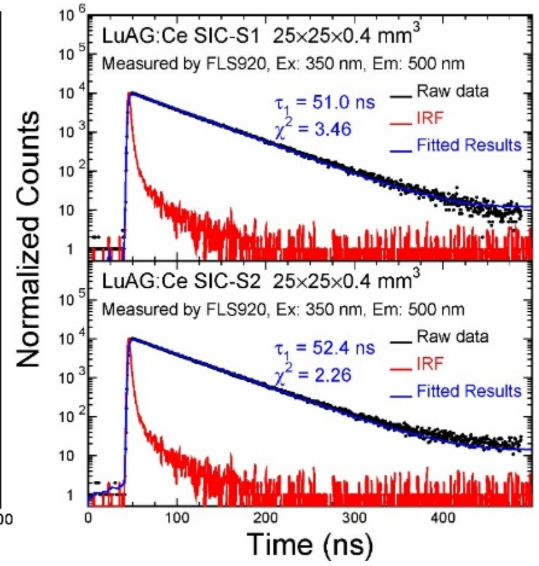
[a] Spurrier, et al., *IEEE T. Nucl. Sci.* 2008,55 (3): 1178-1182
[b] Liu, et al., *Adv. Opt. Mater.*, 4: 731–739. doi: 10.1002/adom.201500691
[c] Yanagida, et al., *IEEE T. Nucl. Sci.* 2012, 59(5): 2146
[d] Luo, et al., *Ceram. Int.* 2016, 41(1): 873
[e] The melting point of these materials various with different composition from 1800 to 1980 °C.
The data is based on reported values.
[f] Kuwano, et al., *J. Cryst. Growth.* 1988, 92: 17
[g] Wu, et al., *NIMA* 2015, 780: 45
[h] Cherepy, et al., *IEEE T. Nucl. Sci.* 2013, 60(3): 2330
[i] van Loef, et al., *IEEE T. Nucl. Sci.* 2007, 54(3):741
[j] Based on ^{137}Cs gamma-ray excitation (shaping time of 4 μs) light yield result in ref. i
[k] Data based on single crystals

Performance of LuAG:Ce Ceramics



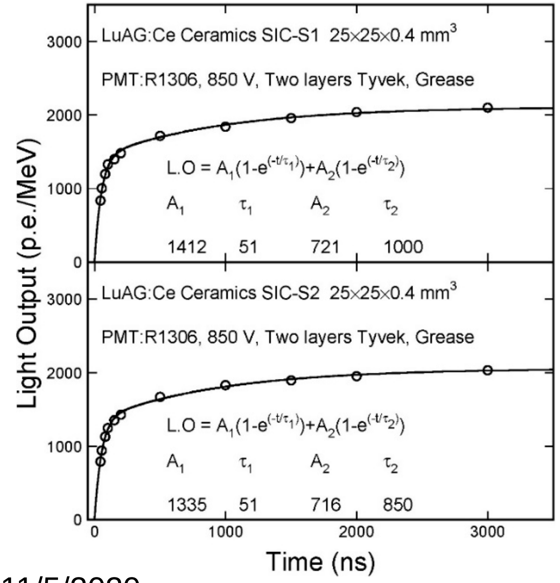


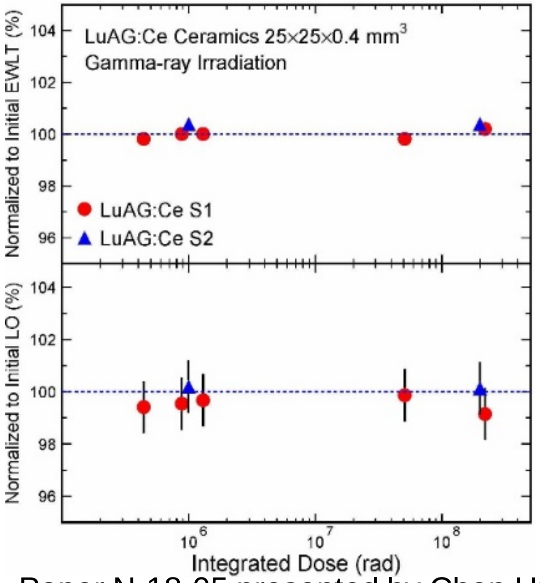


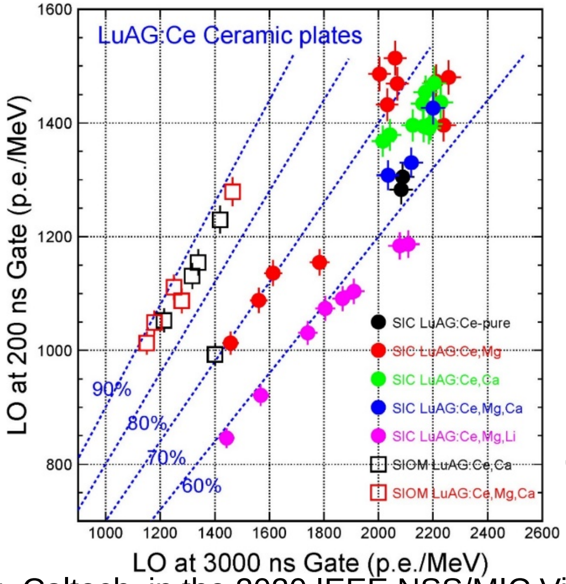


Light output: 1,400 p.e./MeV with a fast decay time of about 50 ns and a slow decay time of about 1 μs. Excellent radiation hardness against ionization dose up to 220 Mrad.

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





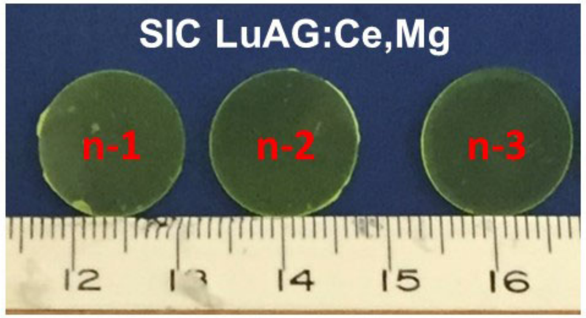


C. Hu, et al., *NIMA* 954 (2020) 161723

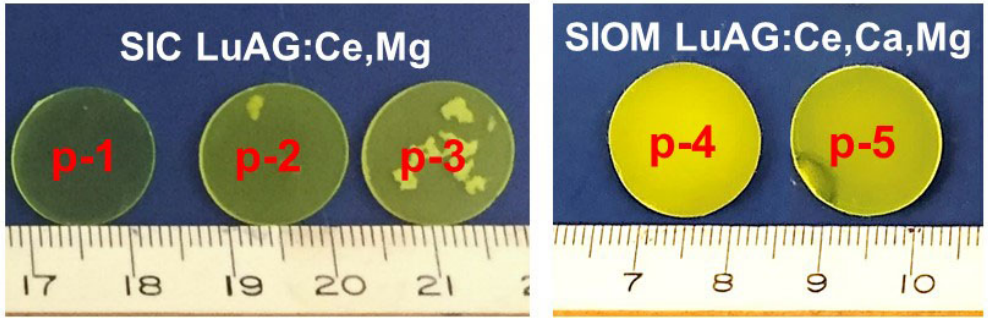
LuAG Samples for Neutron/Proton Irradiation

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



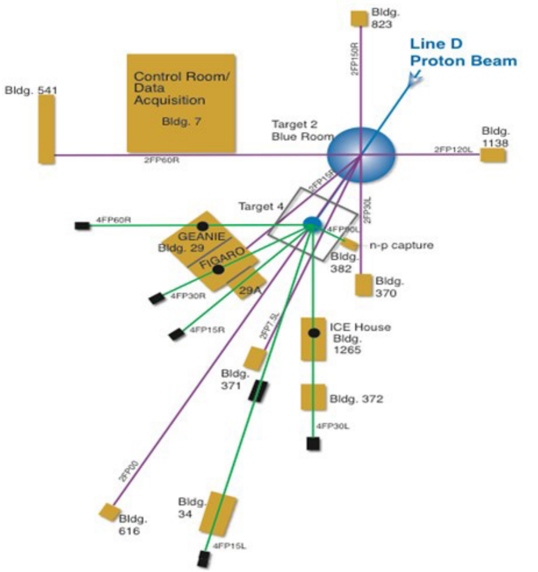
Proton Irradiation Samples

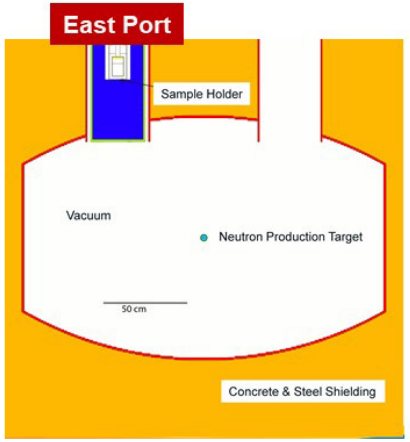


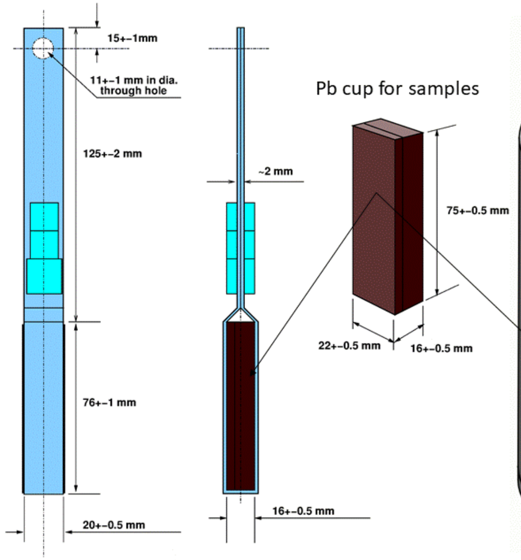
Sample ID	Dimension (mm^3)	200 ns L.O. (p.e./MeV)	F/T ratio (%)	Experiment	Fluence (cm^{-2})
n-1	Φ14.4×1	1474	66.6	LANSCE-7638	1.7×10^{15}
n-2	Φ14.4×1	1479	65.6	LANSCE-7638	3.4×10^{15}
n-3	Φ14.4×1	1514	73.5	LANSCE-7638	6.7×10^{15}

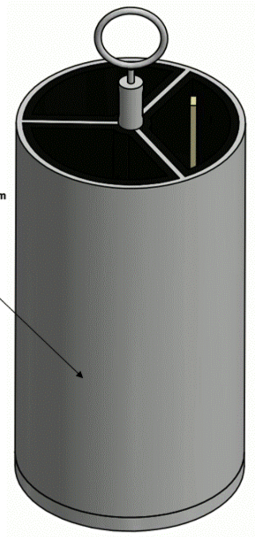
Sample ID	Dimension (mm^3)	200 ns L.O. (p.e./MeV)	F/T ratio (%)	Experiment	Fluence (cm^{-2})
p-1	Φ14.4×1	1486	74.2	CERN	7.1×10^{13}
p-2	Φ14.4×1	1305	62.5	CERN	3.6×10^{14}
p-3	Φ14.4×1	1283	61.6	CERN	1.2×10^{15}
p-4	Φ17×1	1013	88.0	LANSCE-8051	2.4×10^{13}
p-5	Φ17×1	1049	89.0	LANSCE-8051	2.3×10^{14}

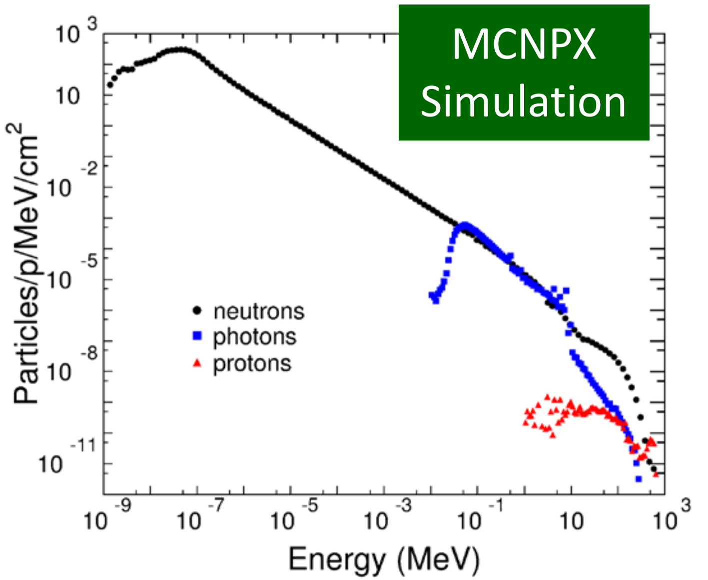
Neutron Irradiation at East Port of LANSCE









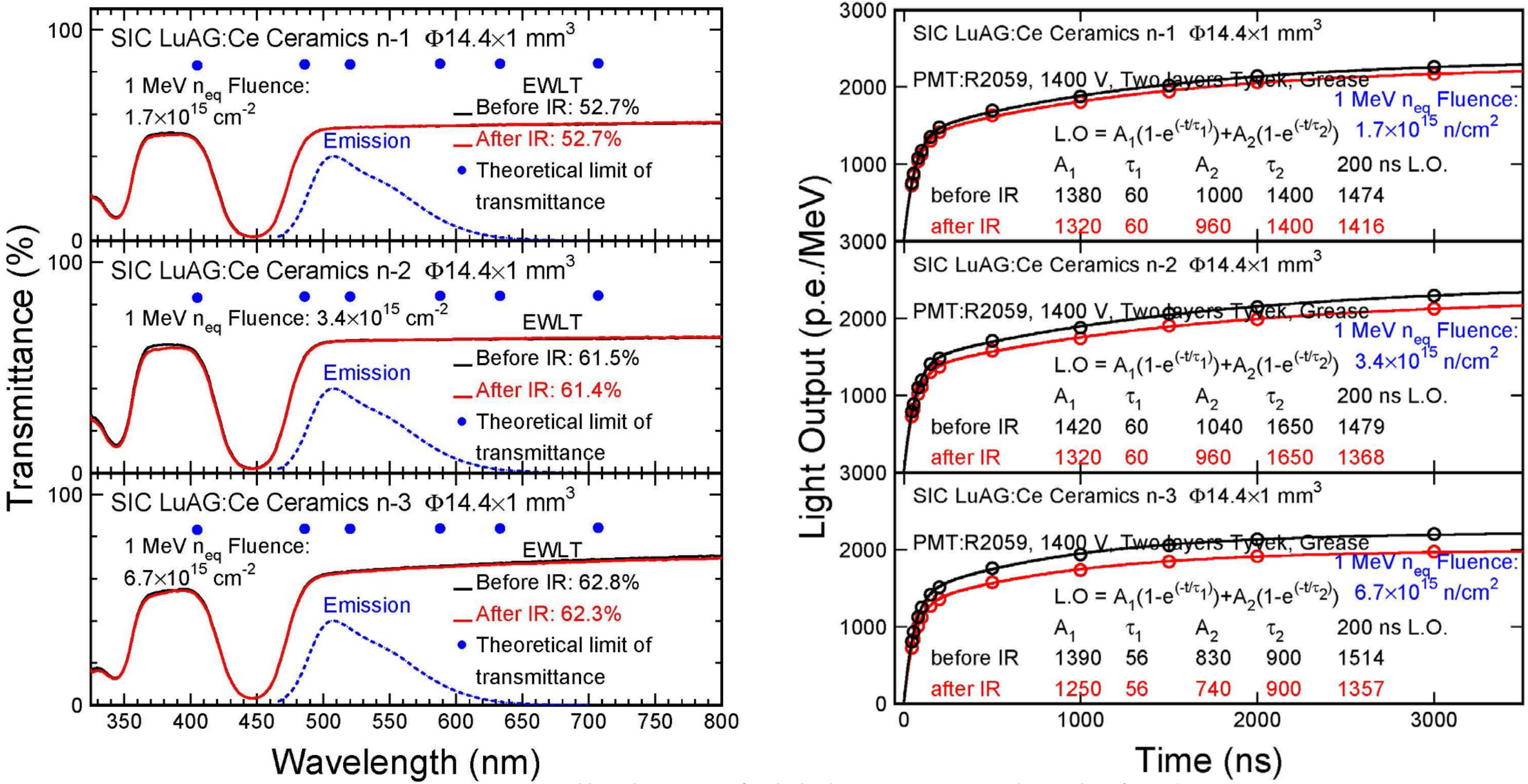


1 MeV equivalent neutron fluence is 1.7, 3.4, and 6.7 $\times 10^{15}$ cm^{-2} for samples n-1, n-2, and n-3, respectively

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}

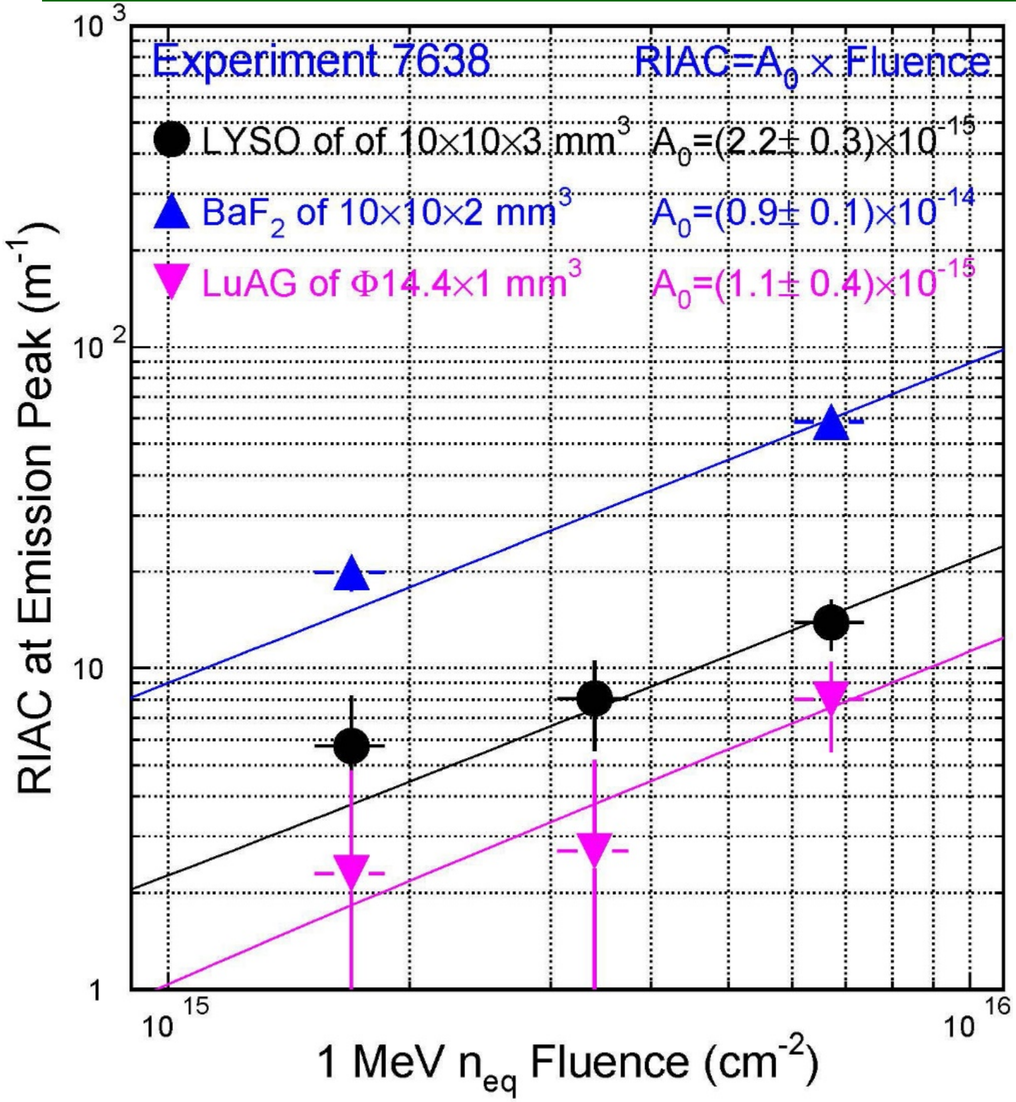
LuAG:Ce after Neutron Irradiations

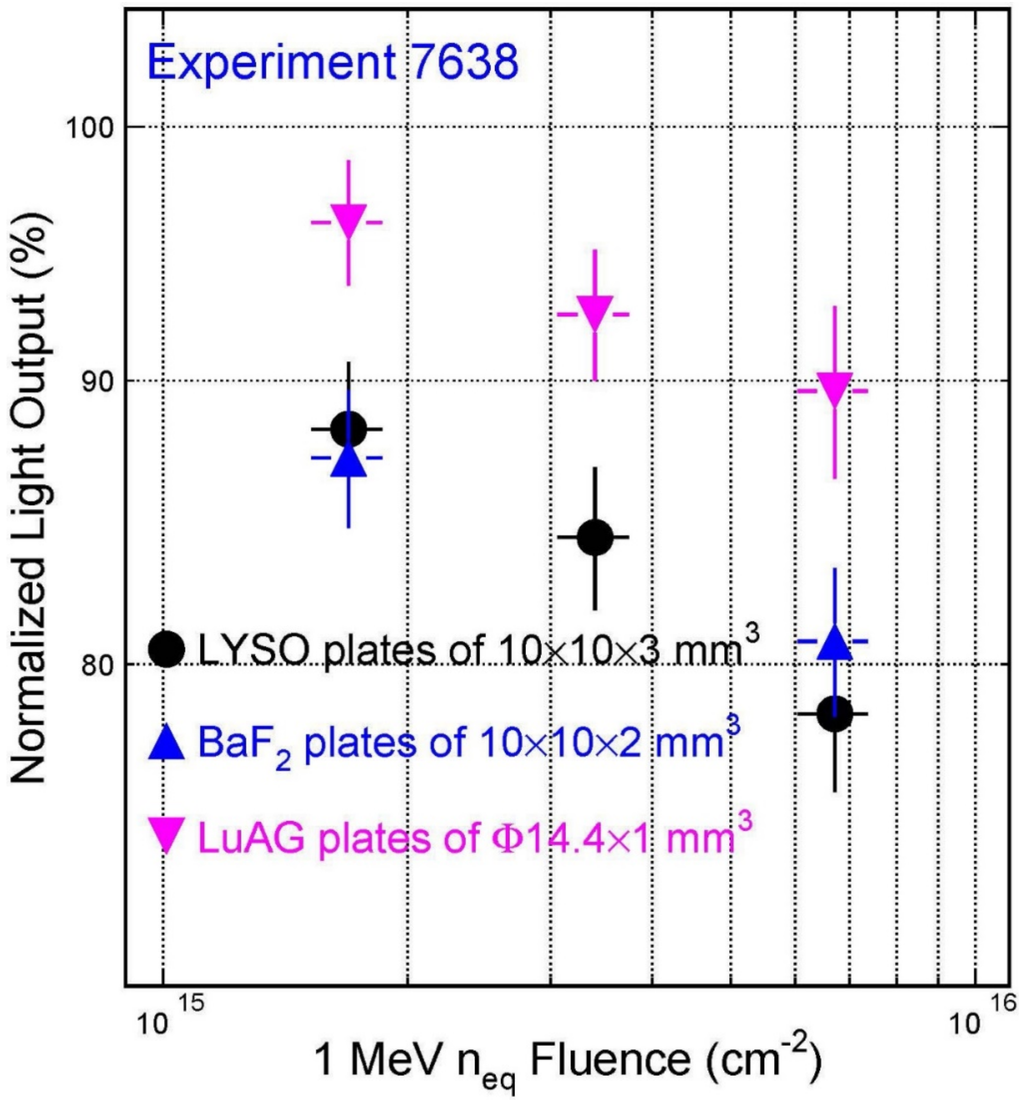
Small losses in T/LO up to 6.7×10^{15} n_{eq}/cm^2 with F/T ratio unchanged



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

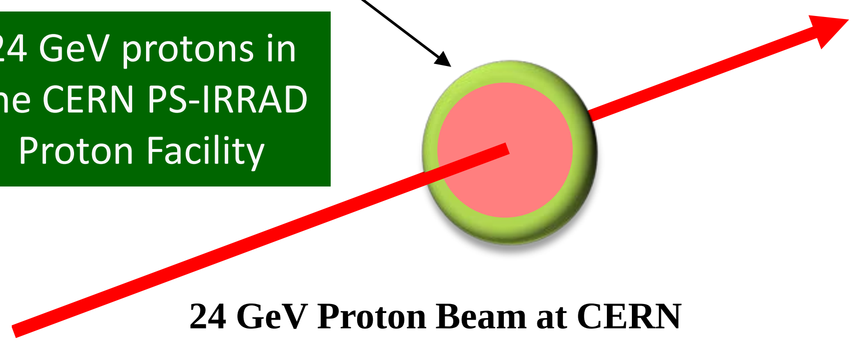




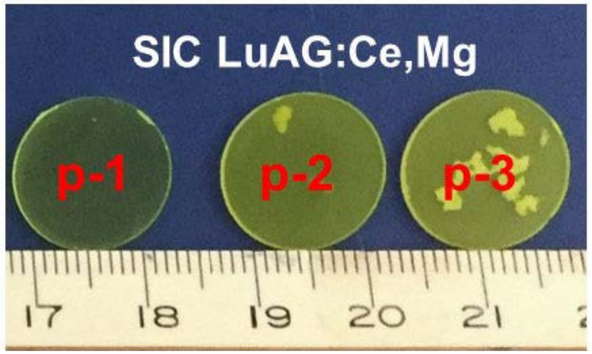
Proton Irradiations at CERN PS & LANSCE

24 GeV protons in the CERN PS-IRRAD Proton Facility

LuAG Plate Φ14.4×1 mm³



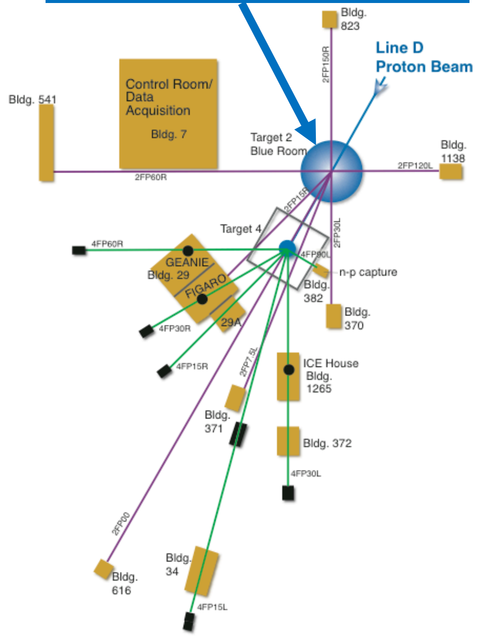
24 GeV Proton Beam at CERN
Gaussian width a FWHM of about 12 mm

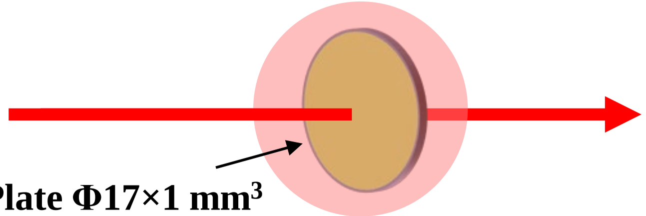


Proton fluence measured by dosimeters of 10×10 and 20×20 mm² at CERN.

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.

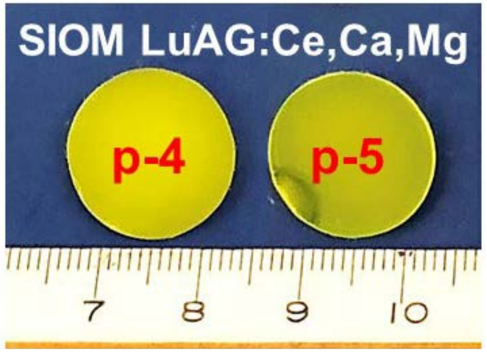
Blue Room in LANSCE





LuAG Plate Φ17×1 mm³

800 MeV proton beam (FWHM= 2.5 cm)

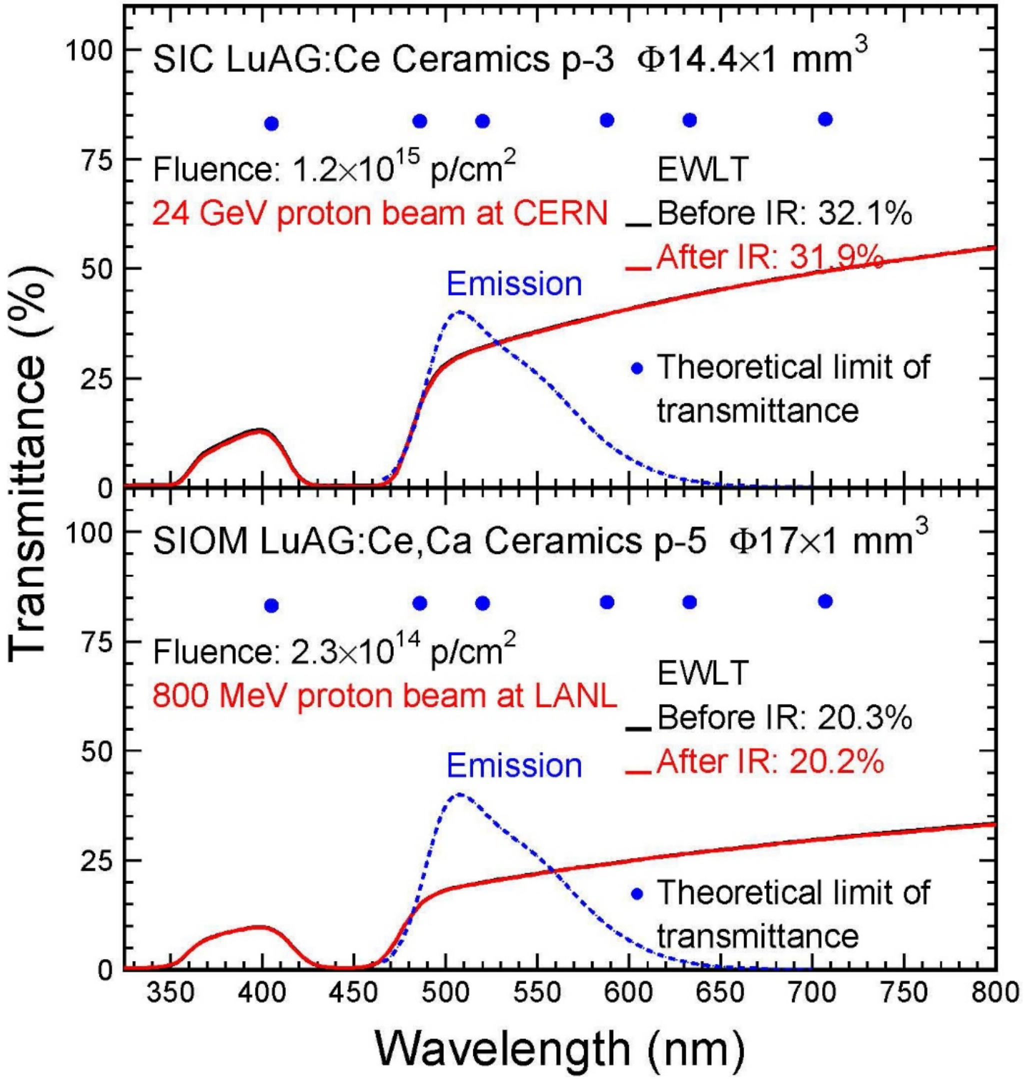


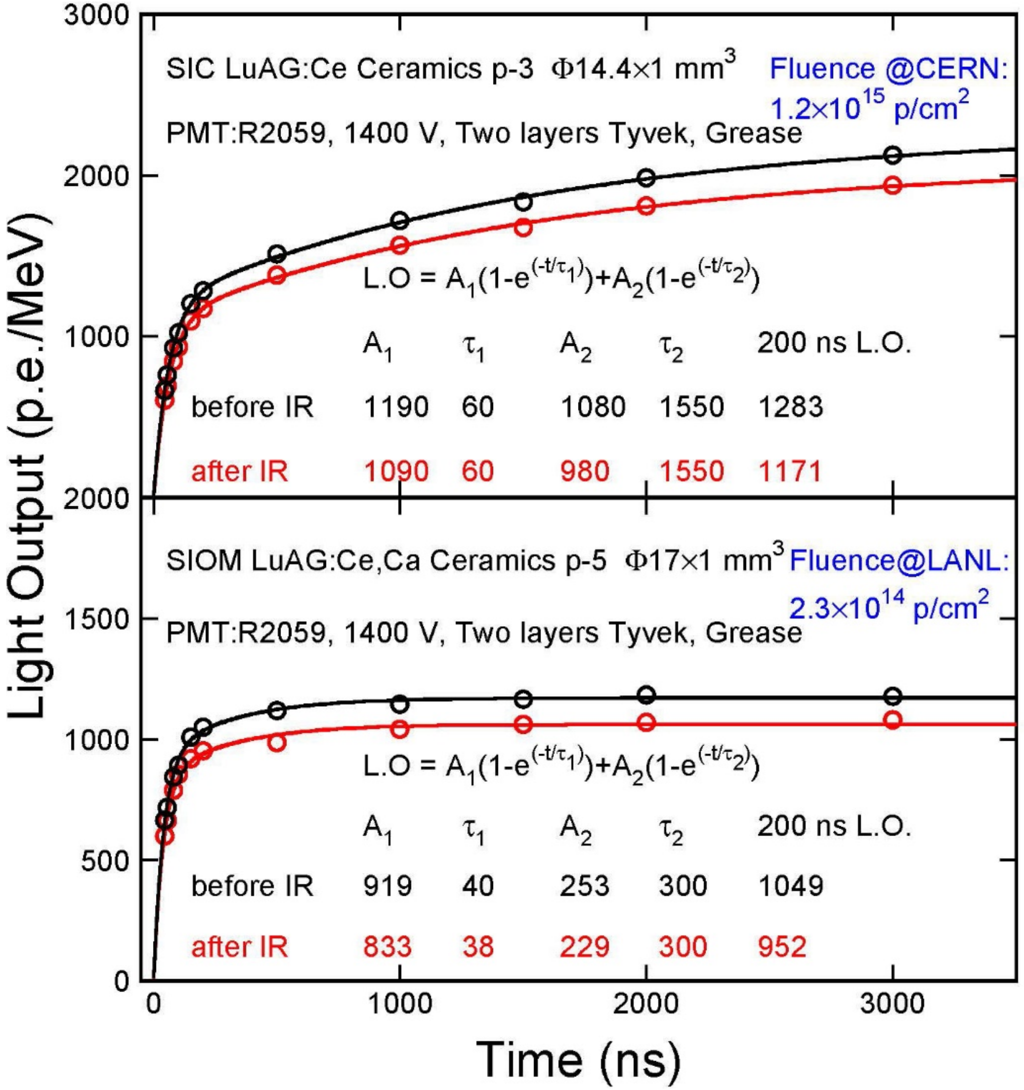
Environment/Source	Proton Flux (p s^{-1} cm^{-2})	Fluence on Crystal (p cm^{-2})
CMS FCAL (η=1.4) at HL-LHC	2.8×10^{5}	2.5×10^{13} / 3000 fb^{-1}
CMS FCAL (η=3.0) at HL-LHC	2.3×10^{6}	2.1×10^{14} / 3000 fb^{-1}
WNR facility of LANSCE	**Up to 2×10^{10}**	**Up to 3×10^{15}**

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

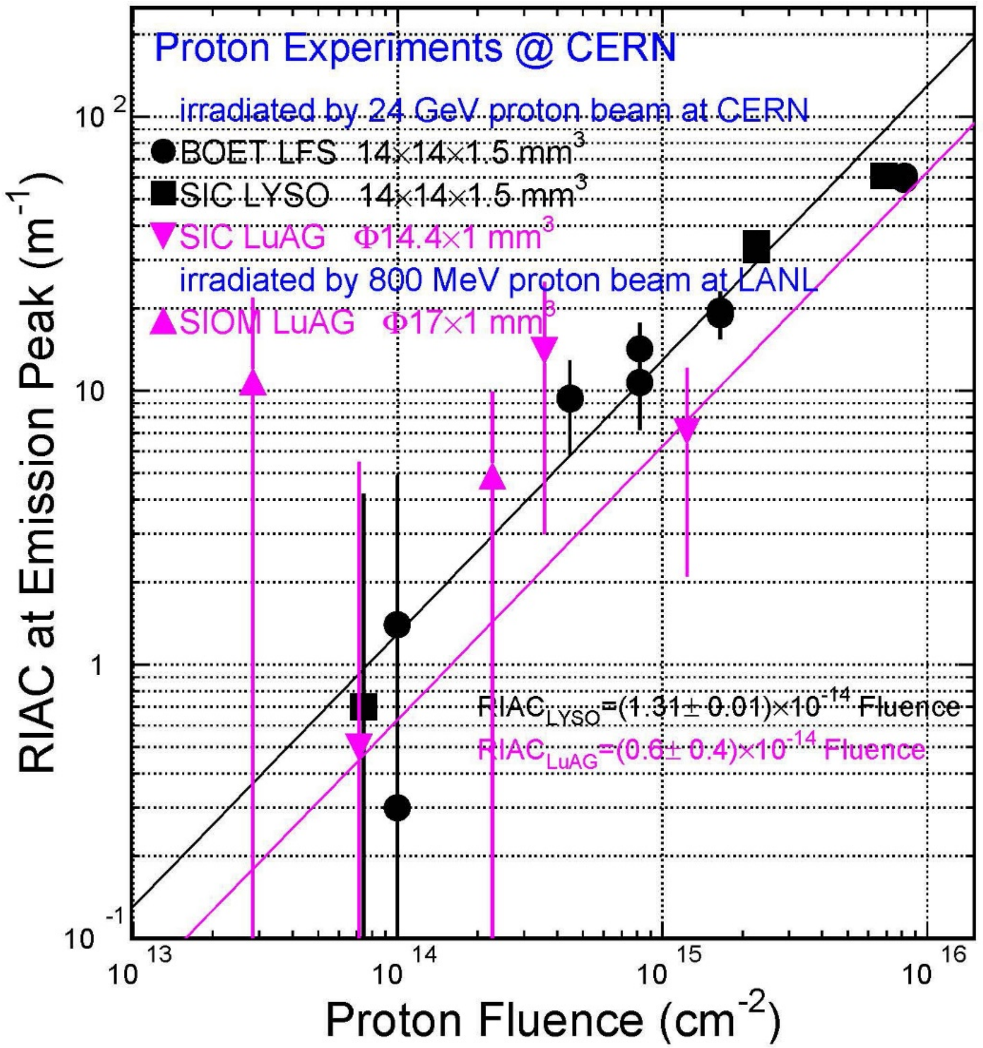
Small loses in T/LO after 1.2×10^{15} p/cm^2 by 24 GeV protons at CERN and after 2.3×10^{14} p/cm^2 by 800 MeV protons at LANSCE with F/T unchanged

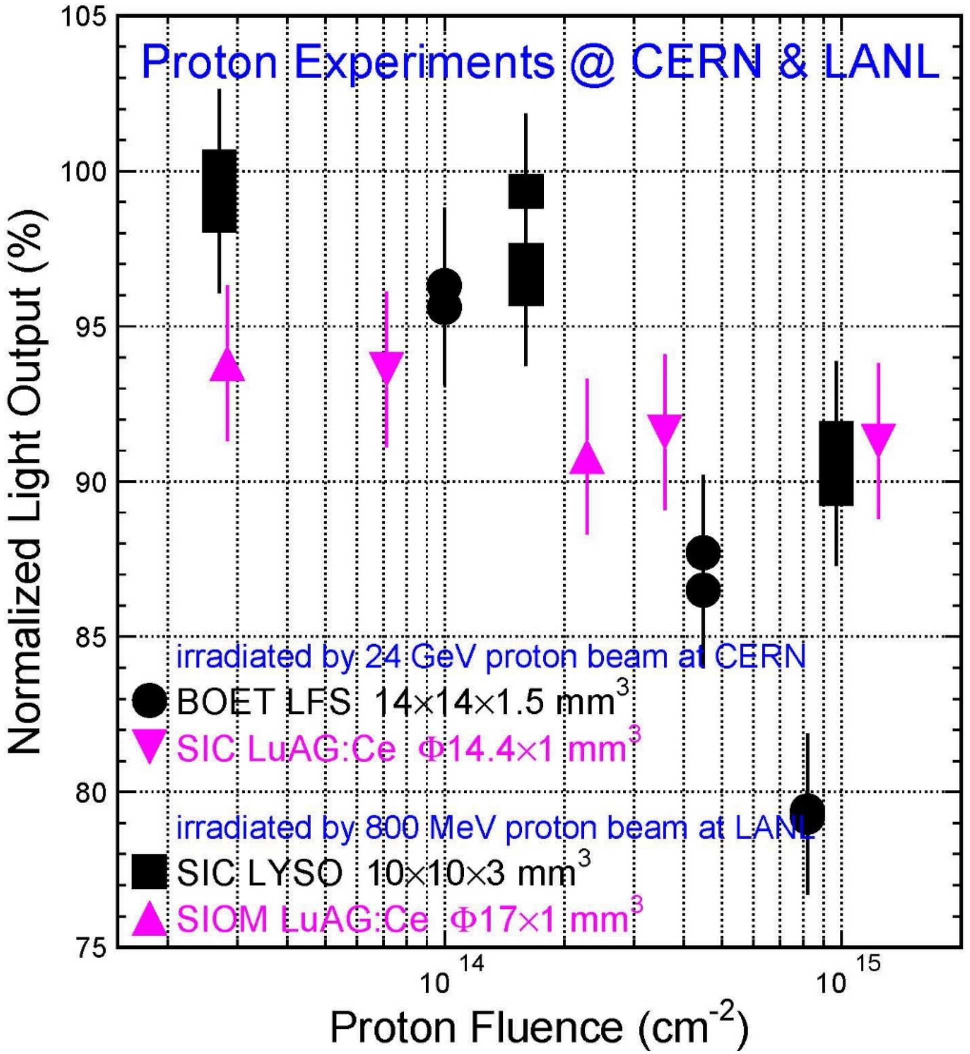




RIAC and LO vs. Proton Fluence

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





Summary

Ca^{2+} and Mg^{2+} co-doped LuAG:Ce ceramic samples were fabricated and irradiated up to 6.7×10^{15} n_{eq}/cm^2 and 1.2×10^{15} p/cm^2 respectively at LANSCE and CERN.

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 will continue to develop Ca^{2+} co-doped LuAG:Ce,Ca ceramics to further improve its optical quality, F/T ratio and radiation hardness.

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