# Inorganic Scintillators for Future HEP Experiments

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# Challenges for Inorganic Scintillators

- **Fast and radiation hard scintillators, such as LYSO:Ce, are needed at the energy frontier, e.g. CMS ECAL Endcap (η=3) at HL-LHC with 3000 fb$^{-1}$:**
  - **Absorbed dose: up to 60 Mrad (dose rate 5 krad/hr),**
  - **Charged hadron fluence: up to $6\times10^{14}$ p/cm$^2$,**
  - **Fast neutron fluence: up to $3\times10^{15}$ n/cm$^2$.**
- **Ultra-fast scintillators, such as $BaF_2$ with sub-ns decay time and suppressed slow scintillation component, are needed to face the challenge of unprecedented event rate expected at the intensity frontier, e.g. Mu2e-II.**
- **Cost-effective scintillators are needed for a homogeneous hadronic calorimeter (HHCAL) detector concept to achieve excellent jet mass resolution at future high energy lepton colliders, such as ILC, CLIC, FCC and CEPC.**

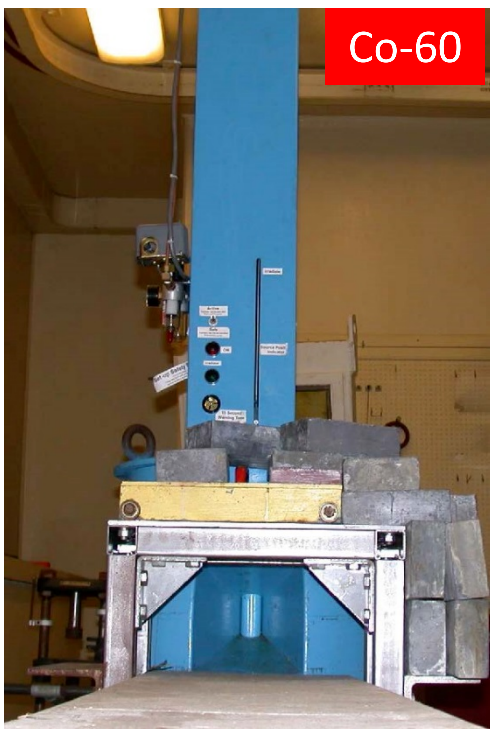
# Bright, Fast Scintillators: LSO/LYSO

| Crystal | NaI(Tl) | CsI(Tl) | CsI | $BaF_2$ | 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<br>220 | 480 | 402 | 425<br>420 | ? |
| Decay Time [b] (ns) | 245 | 1220 | 26 | 650<br>0.9 | 300 | 40 | 30<br>10 | ? |
| Light Yield [b,c] (%) | 100 | 165 | 4.7 | 36<br>4.1 | 21 | 85 | 0.3<br>0.1 | ? |
| d(LY)/dT [b] (%/ °C) | -0.2 | 0.4 | -1.4 | -1.9<br>0.1 | -0.9 | -0.2 | -2.5 | ? |
| Experiment | Crystal Ball | BaBar<br>BELLE<br>BES III | KTeV<br>S.BELLE<br>Mu2e | (GEM)<br>TAPS<br>Mu2e-II? | L3<br>BELLE<br>HHCAL? | COMET<br>(CMS, Mu2e, SuperB) | CMS<br>ALICE<br>PANDA | A4<br>g-2<br>HHCAL? |

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

# Gamma Irradiations

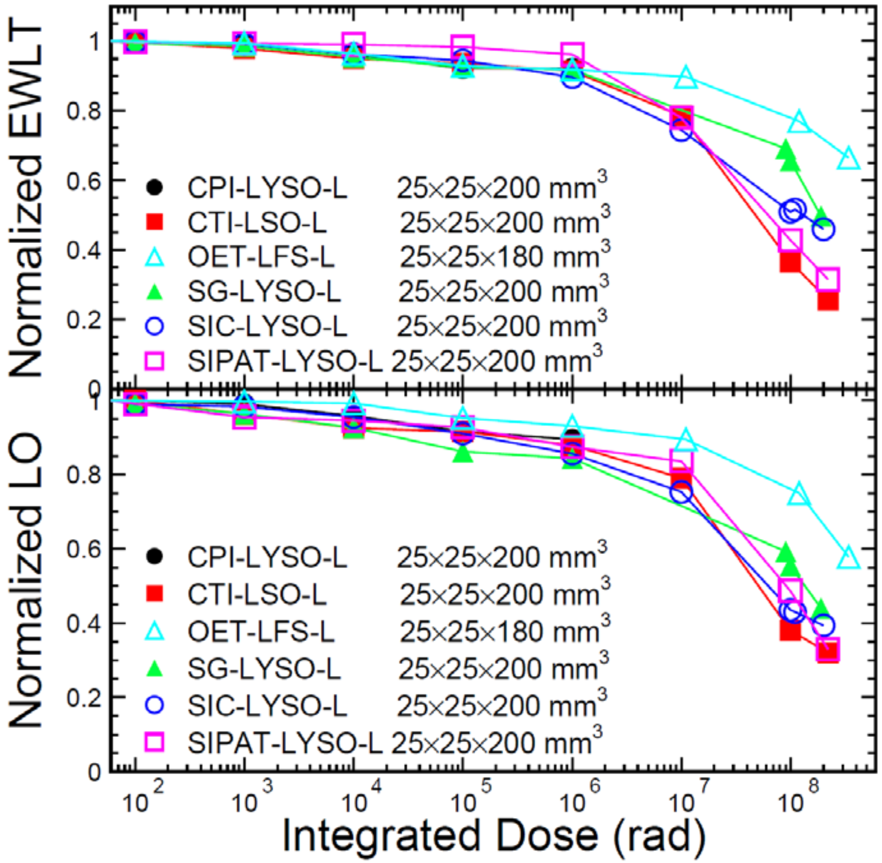
- Crystal samples of about 20 cm long were irradiated step by step at Caltech, JPL and Sandia facilities by $^{60}$Co and $^{137}$Cs sources up to 340 Mrad with a dose rate up to 1 Mrad/h.
- Longitudinal Transmittance (LT) and Light Output (LO) were measured at room temperature before and after each irradiation step within 1 hr.

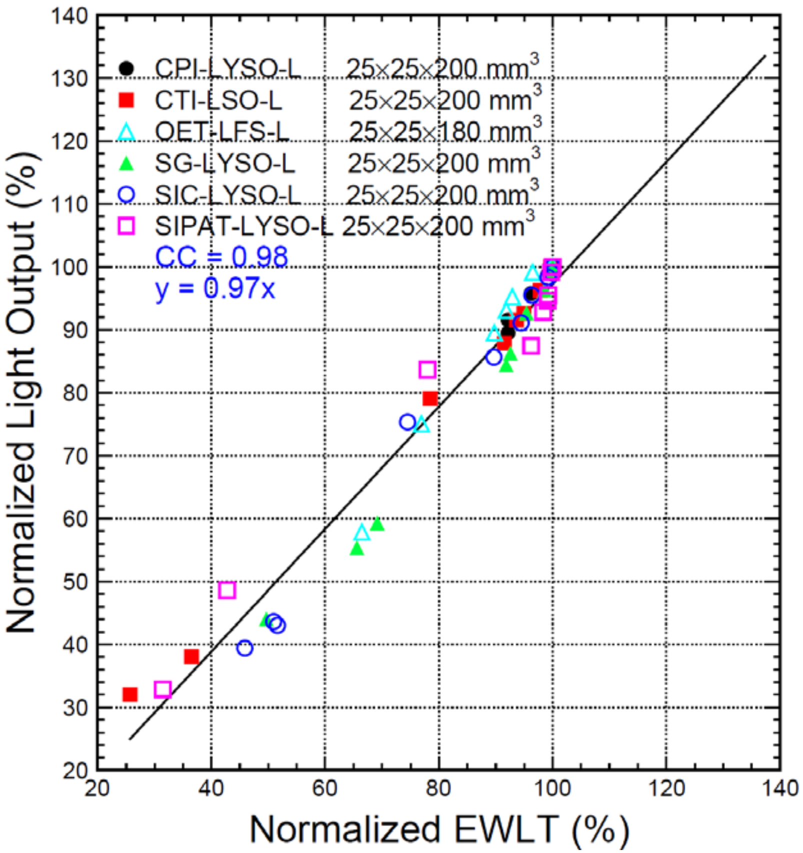




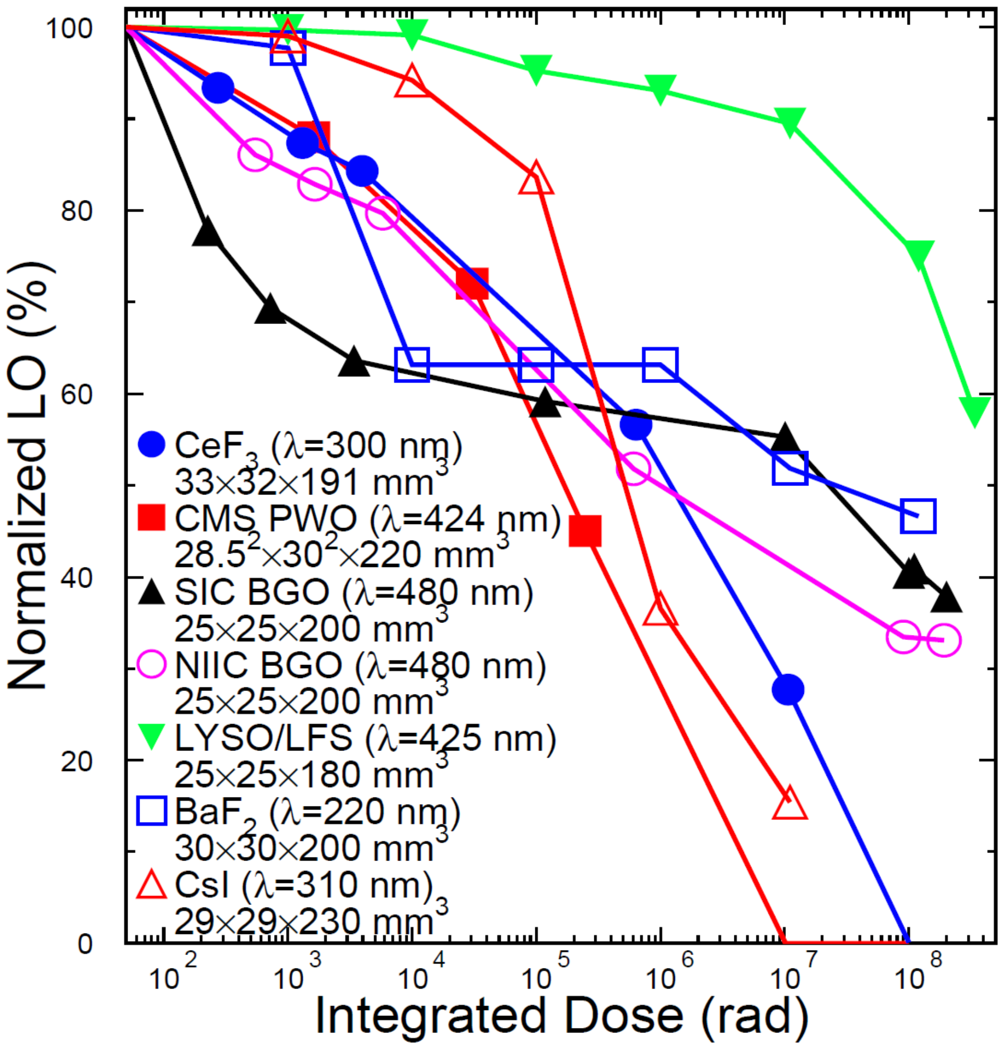
# 20 cm Long LYSO: Normalized EWLT and LO vs. Dose and LO vs. EWLT

- LYSO/LSO/LFS were found the hardest ones against γ irradiation.
- The best sample: 58% light output after 340 Mrad.
- Good correlation between LO and EWLT: LO loss is caused by absorption.



# All Long Crystals: LO Loss

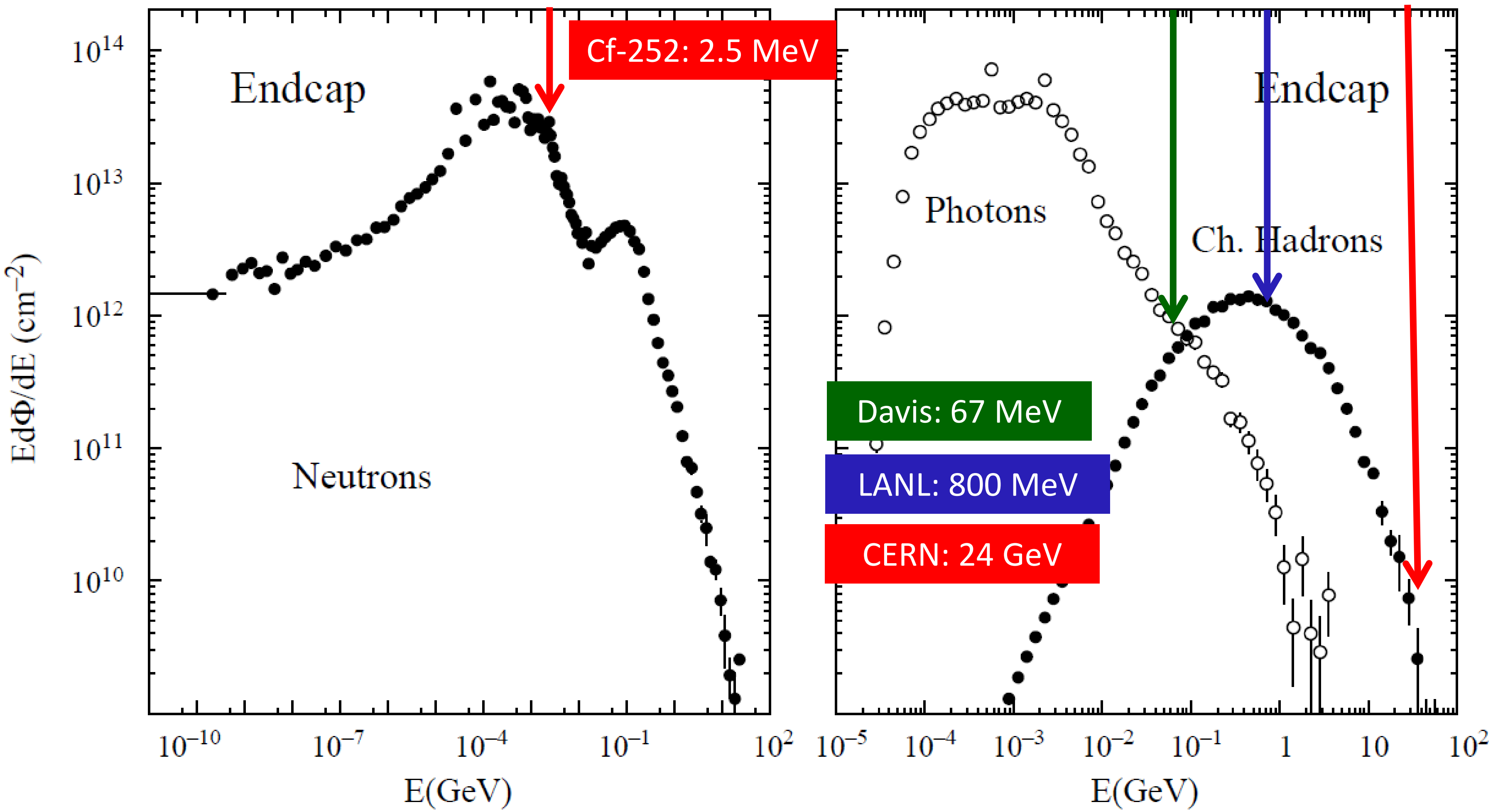



Ignoring dose rate dependence, normalized light output shown as a function of the integrated dose.

LYSO crystals show the best radiation hardness up to 340 Mrad

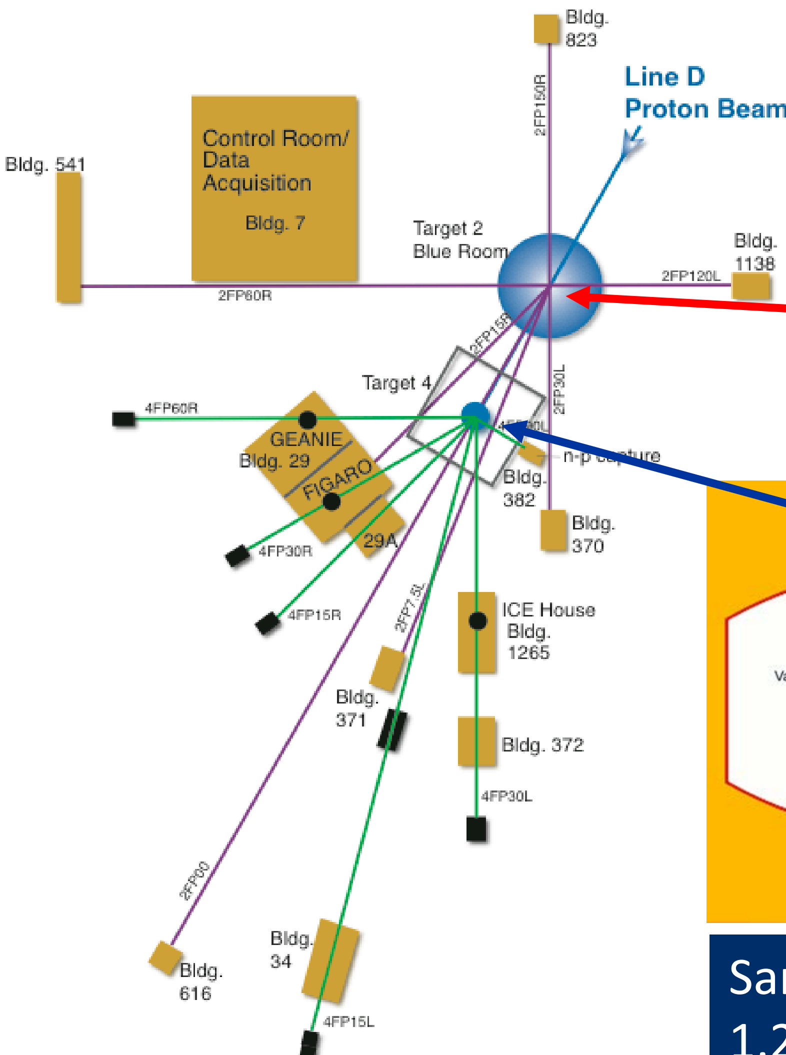
# Particle Energy Spectra at HL-LHC

FLUKA simulations: neutrons and charged hadrons are peaked at MeV and several hundreds MeV respectively. Neutron energy of 2.5 MeV from Cf-252 source and proton energy of 800 MeV at LANL are ideal for such investigation



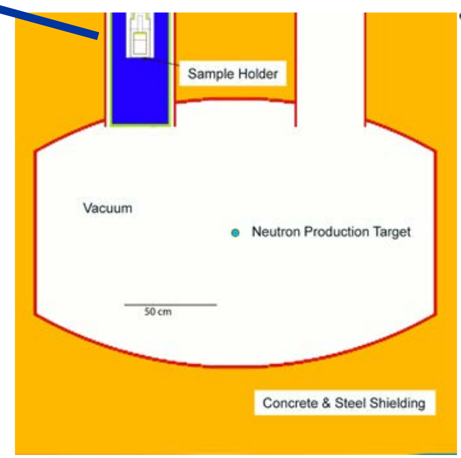
# Proton/Neutron Irradiation at LANL

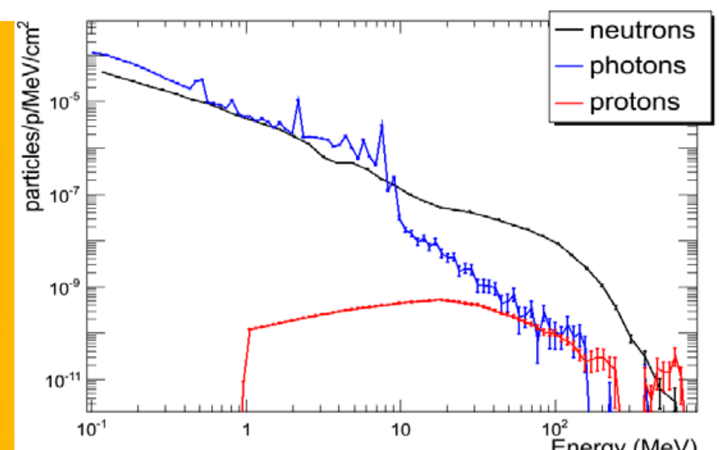
**Los Alamos Neutron Science Center (LANSCE)**




| Environment/Source | Proton Flux ($p\ s^{-1}\ cm^{-2}$) | Fluence on Crystal ($p\ cm^{-2}$) |
|---|---|---|
| CMS FCAL ($\eta$=1.4) at HL-LHC | $4.0 \times 10^4$ | $2.4 \times 10^{12}$ / 3000 $fb^{-1}$ |
| CMS FCAL ($\eta$=3.0) at HL-LHC | $5.0 \times 10^6$ | $3.0 \times 10^{14}$ / 3000 $fb^{-1}$ |
| **WNR facility of LANSCE** | **Up to $2 \times 10^{10}$** | **Up to $3 \times 10^{15}$** |

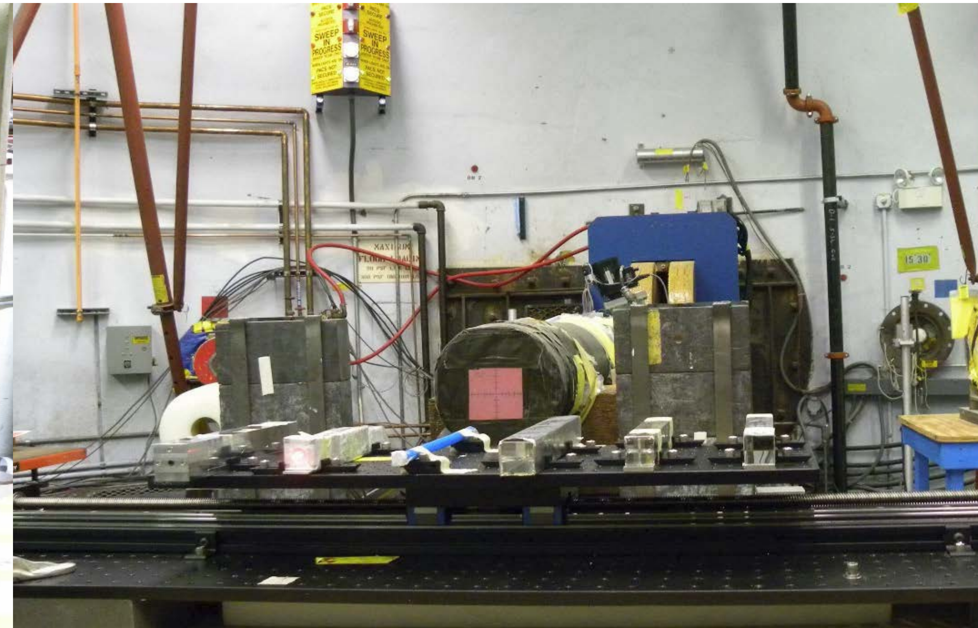
800 MeV proton beam (FWHM= 2.5 cm)

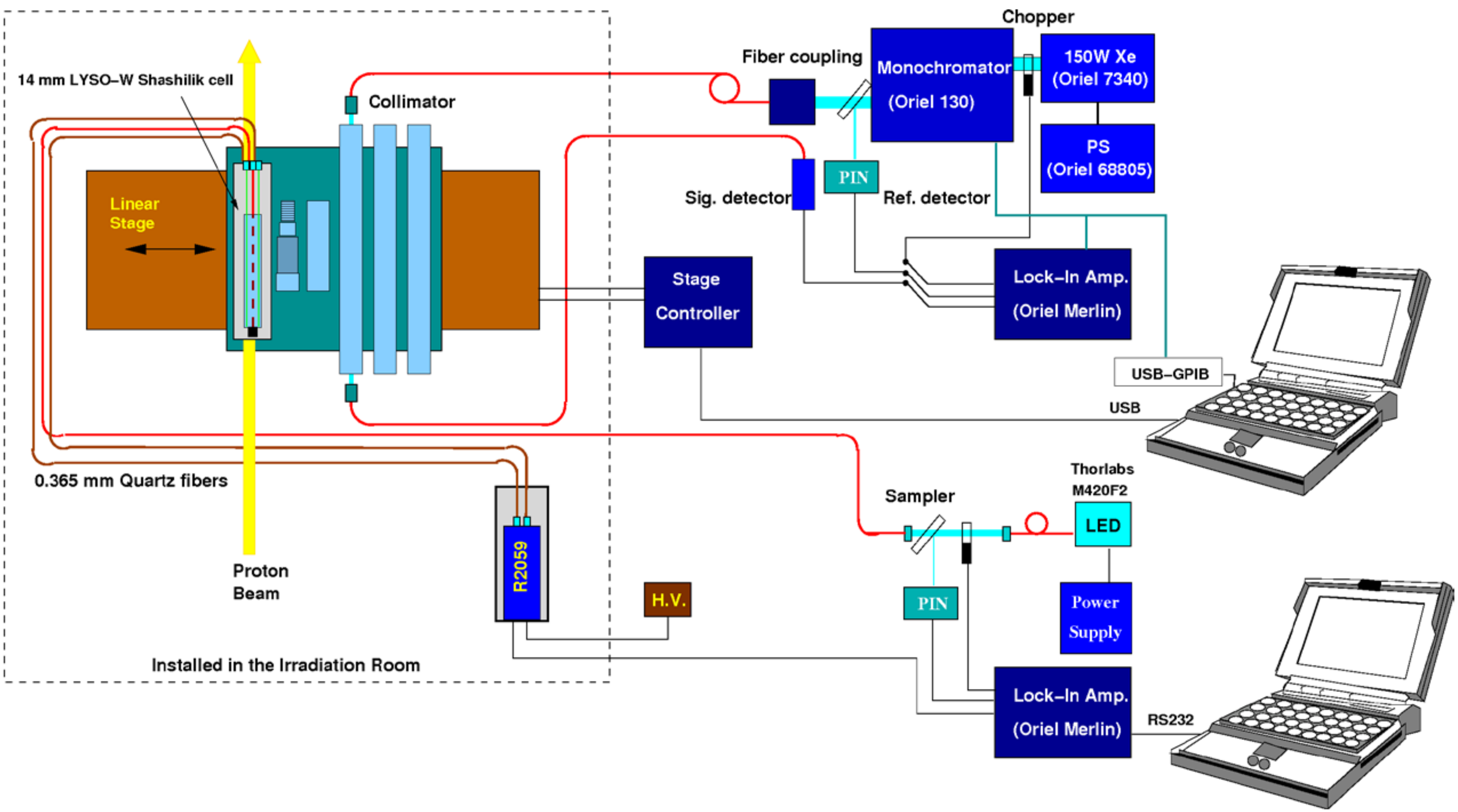

**Samples are placed at Target-4 East Port, about 1.2 m away from the neutron production target.**

# 800 MeV p-Irradiation at LANL

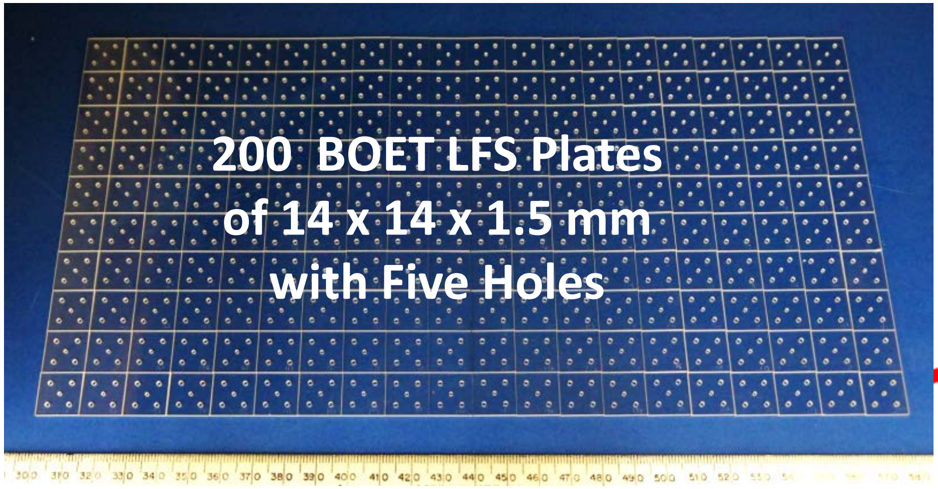



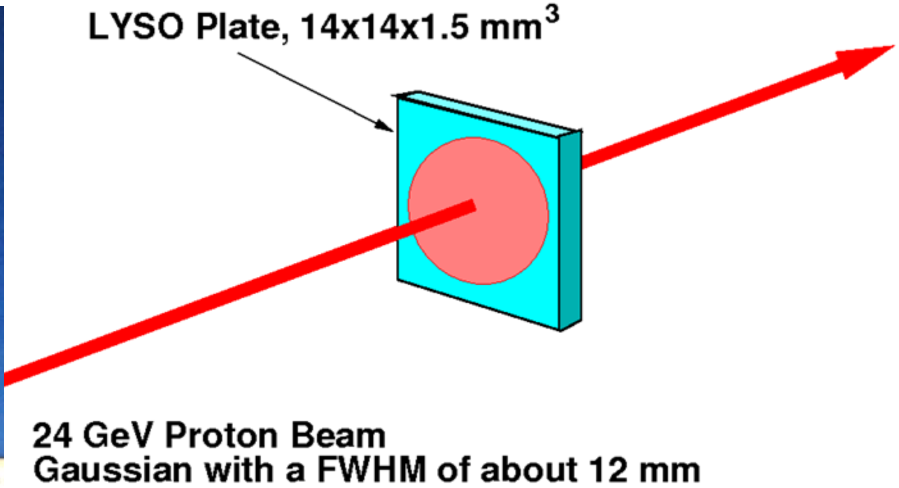
# The 2015 p-Irradiation Setup

- LT (300-800 nm) of long crystals was measured before and after each irradiation step by a Xenon lamp and fiber based spectrophotometer.
- A LYSO-W-Capillary Shashlik cell was monitored before and after each irradiation step by a 420 LED based monitoring system.



# Proton Irradiation for LYSO Plates
# 24 GeV at CERN, 67 MeV at UC Davis




## CERN 24 GeV Proton Irradiation with a beam FWHM of 12 mm

Thanks to David Bailleux and Federico Ravotti

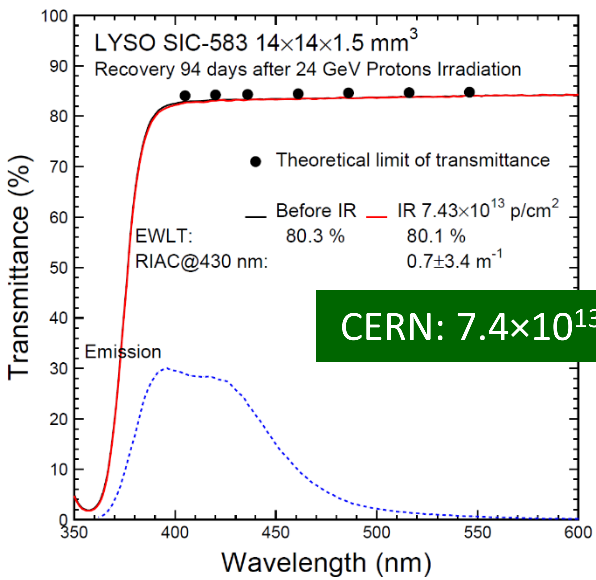
- Four LYSO plates: $7.4\times10^{13}$, 2x $2.4\times10^{15}$ and $6.9\times10^{15}$ p/cm$^2$ in 2014
- Ten LFS plates were irradiated as five pairs from $9.97 \times 10^{13}$ up to $8.19 \times 10^{15}$ p/cm$^2$ in 2015.
- Transmittance and light output were measured after cooled down.

## UC Davis 67 MeV proton irradiation with a beam FWHM of 25 mm

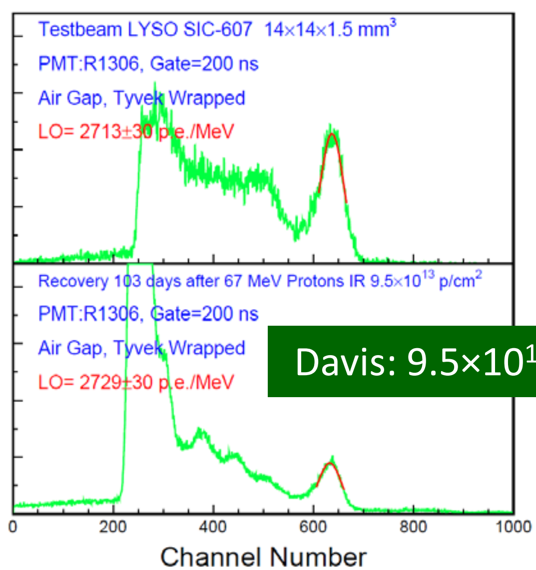
Thanks to Bob Hirosky and Mike Mulhearn

- Five LYSO plates: 2x $1.2\times10^{12}$, $1.2\times10^{13}$, $2.2\times10^{13}$ and $9.5\times10^{13}$ p/cm$^2$

# T and LO Losses after p-Irradiation

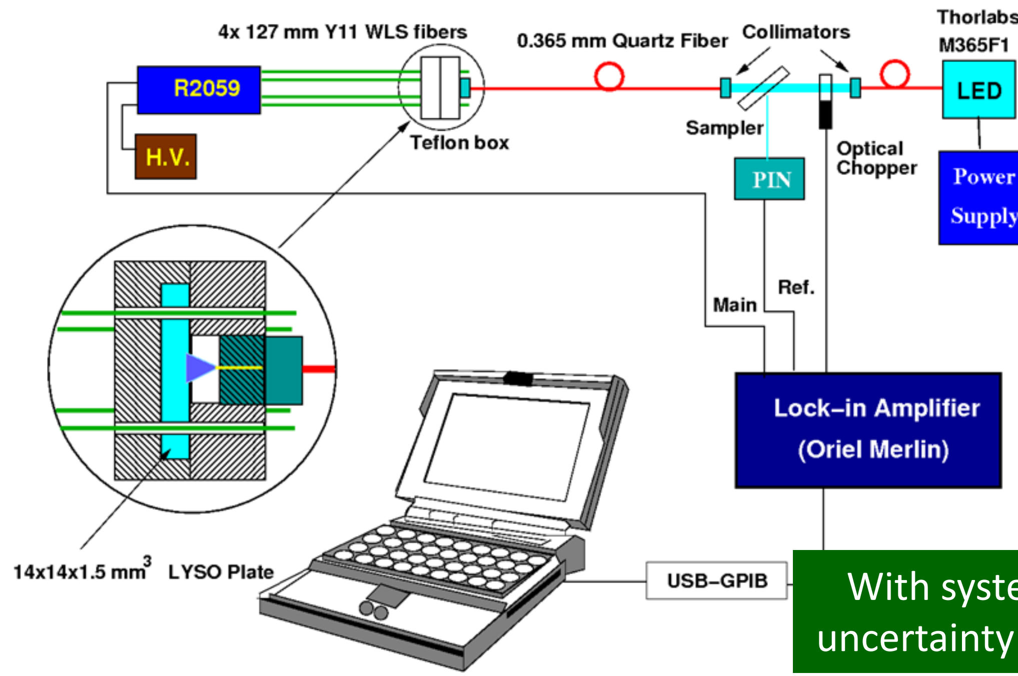



CERN: $7.4\times10^{13}$




Davis: $9.5\times10^{13}$

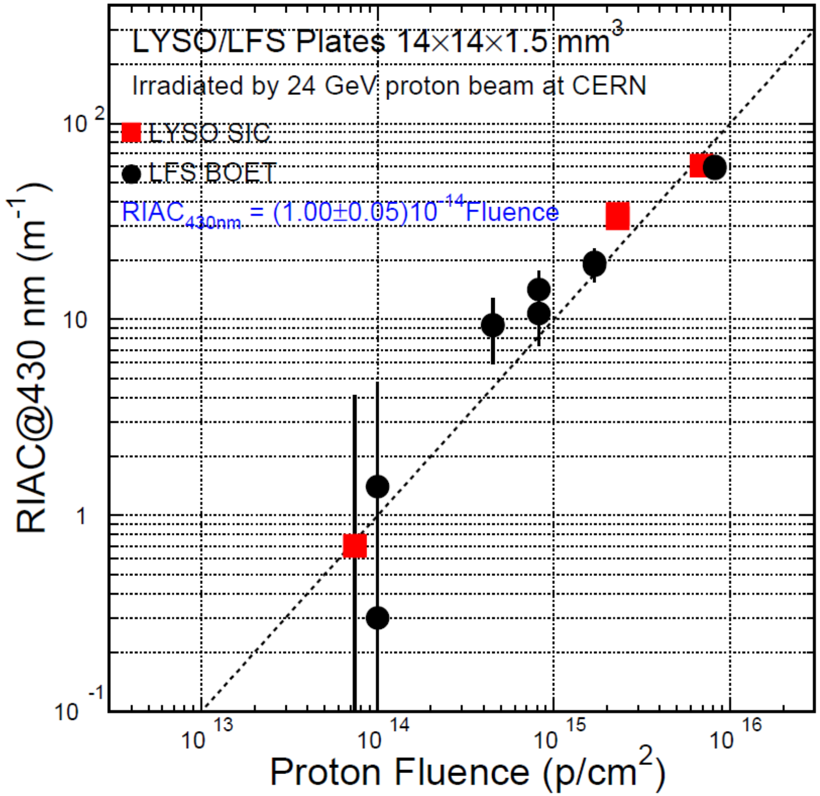
- Transmittance measured by a PerkinElmer Lambda 950 spectrophotometer with 0.15% precision or 3.5/m for radiation induced absorption (RIAC).
- LO measured with PMT via direct coupling or through 4 x Y11 WLS fibers to mitigate phosphorescence.
- No loss was observed up to $10^{14}$ p/cm$^2$ for 67 MeV and 24 GeV protons.

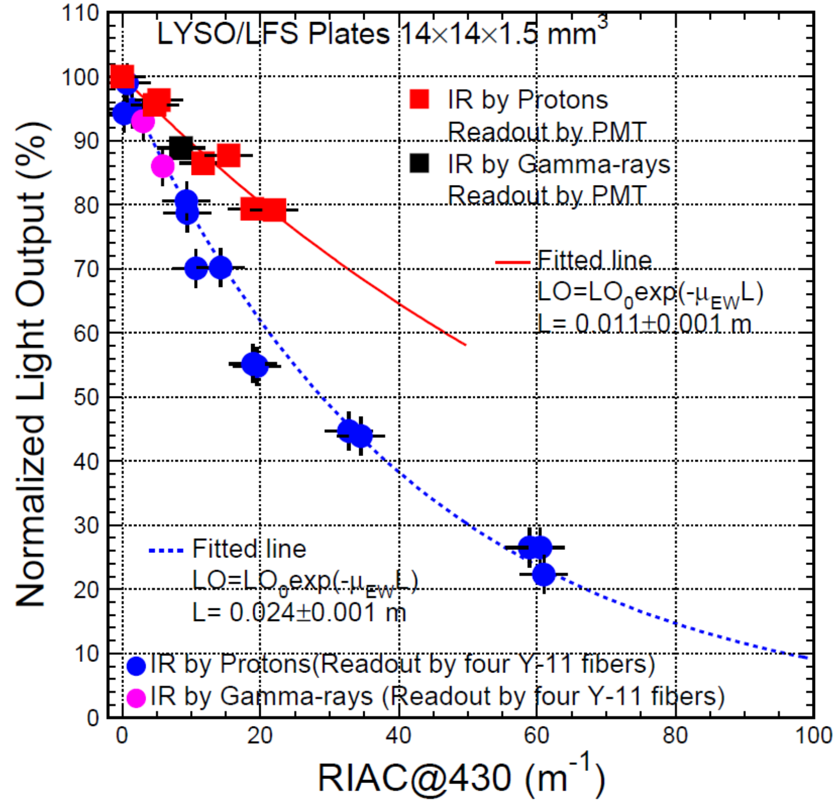



With systematic uncertainty of 2.5%

# RIAC and LO after p-Irradiation

- Consistent RIAC at 430 nm is observed in LYSO and LFS plates irradiated by 24 GeV protons up to 8.19 x $10^{15}$ p/cm$^2$ at CERN in 2014 and 2015.
- Damages by protons and gamma-rays are consistent in terms of LO vs. RIAC, indicating that LO loss is caused by absorption so can be monitored.
- Average light path length of 1.1 and 2.4 cm at 430 nm for direct and Y-11 readout.



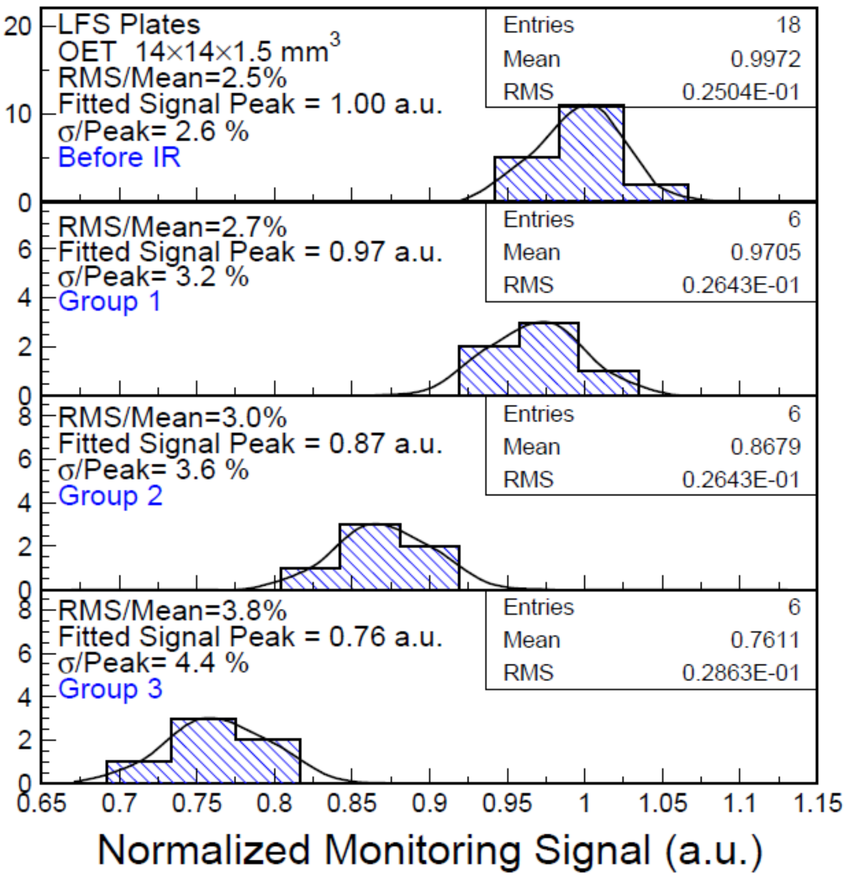
# Neutron Irradiation 2015 at LANL

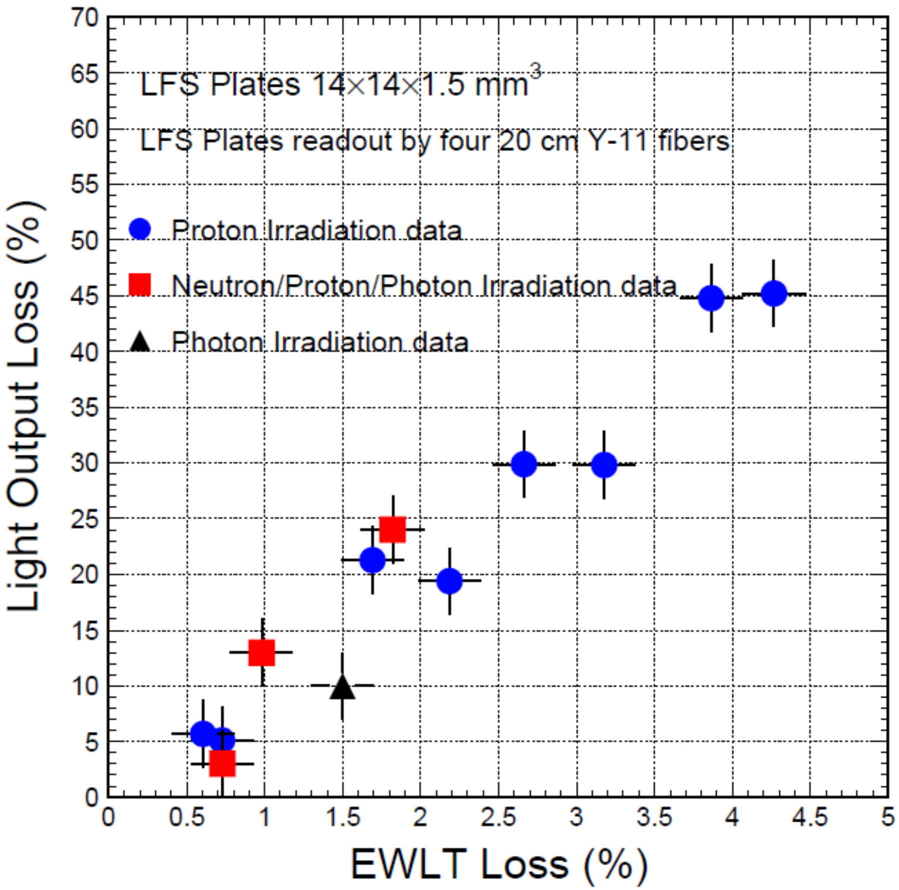
- A total of 18 LFS plates of 14×14×1.5 mm$^3$ were divided into 3 groups and removed after 13.4, 54.5 and 118 days respectively.
- Samples were shipped back after cooling down and with light output and transmittance measured at Caltech.

| Particles / Dose | Group-1 (BOET 107-112) Fluence (cm$^{-2}$) | Group-2 (BOET 101-106) Fluence (cm$^{-2}$) | Group-3 (BOET 95-100) Fluence (cm$^{-2}$) |
|---|---|---|---|
| Thermal and Epithermal, Neutrons (0<En < 1 eV) | 7.01E+14 | 3.16E+15 | 5.64E+15 |
| Slow and Intermediate Neutrons (1 eV <En < 1 MeV) | 2.56E+15 | 1.15E+16 | 2.05E+16 |
| Fast Neutrons (En > 1 MeV) | 2.24E+14 | 1.01E+15 | 1.80E+15 |
| Protons (Ep>1 MeV) | 5.31E+11 | 2.39E+12 | 4.27E+12 |
| Protons Ionization Dose (rad) | 1.39E+04 | 6.25E+04 | 1.12E+05 |
| Photons (Eg>150 KeV) | 6.71E+14 | 3.02E+15 | 5.39E+15 |
| Photons Ionization Dose (rad) in Air | 2.40E+07 | 1.08E+08 | 1.93E+08 |

# LO and EWLT after n-Irradiation

- ➢ Light output was measured by UV LED excitation and Y-11 WLS fibers, see slide 12.
- ➢ Degradations of 3%,13% and 24% is observed for Group-1, 2 and 3 respectively.
- ➢ LO loss vs. EWLT loss is consistent with that of gamma and proton irradiated ones.
- ➢ Pb shielding is implemented in 2016 irradiation to evaluate gamma contribution.



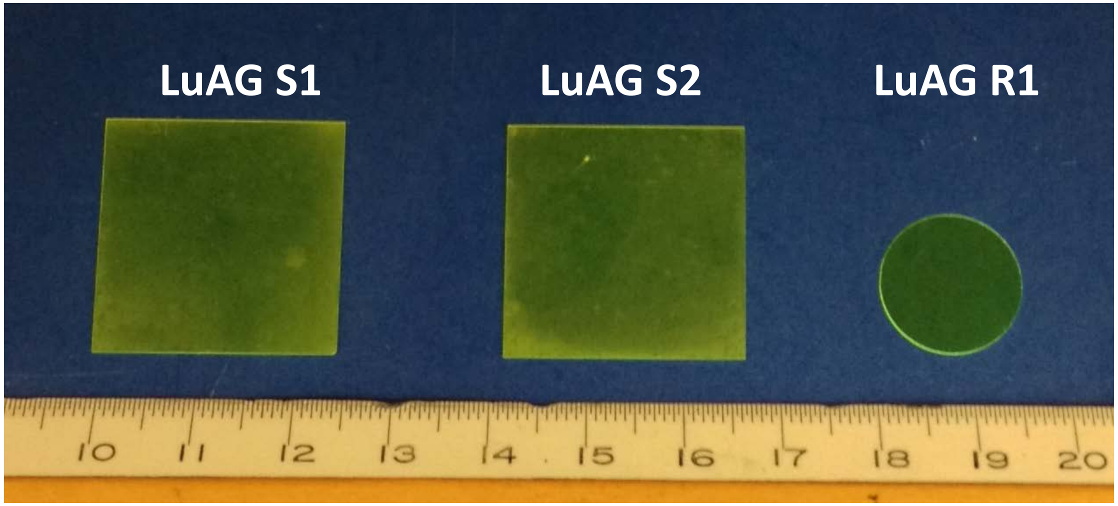



# Inorganic Ceramic Scintillators

| | 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 |
| $\lambda_I$ (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 |
| $\lambda_{peak}$ (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<br>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

# LuAG:Ce Samples from SIC



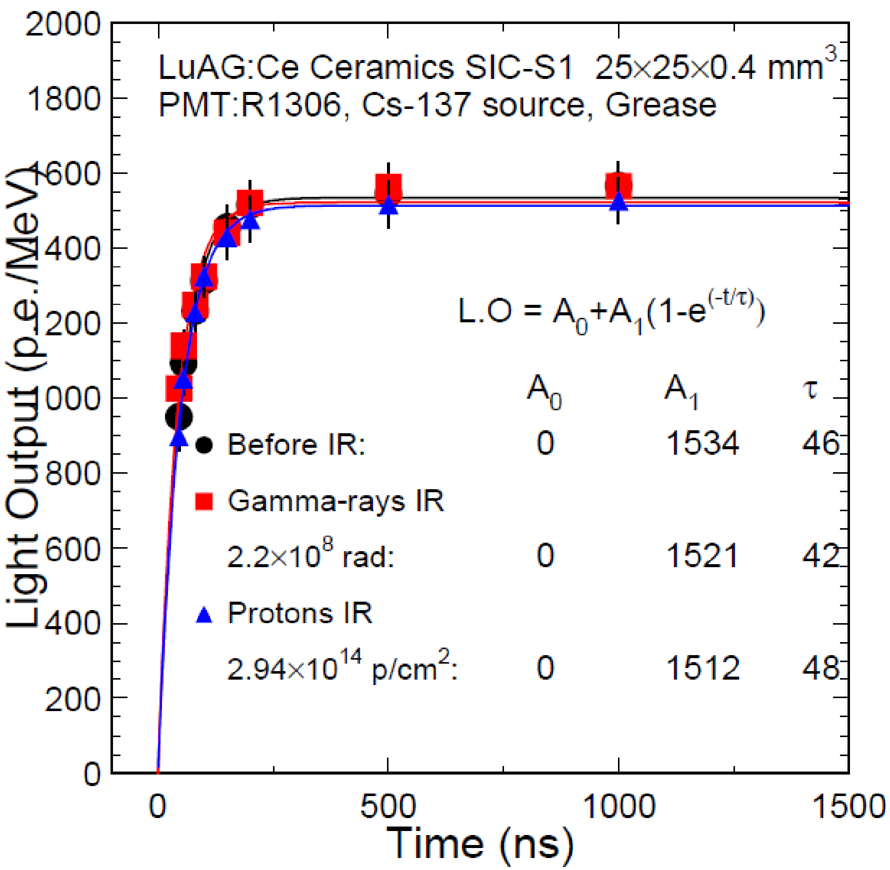

| Sample ID | Dimension (mm) | Polishing |
|---|---|---|
| LuAG S1 | 25×25×0.4 | Two surfaces |
| LuAG S2 | 25×25×0.4 | Two surfaces |
| LuAG RI | Φ15×0.2 | Two surfaces |

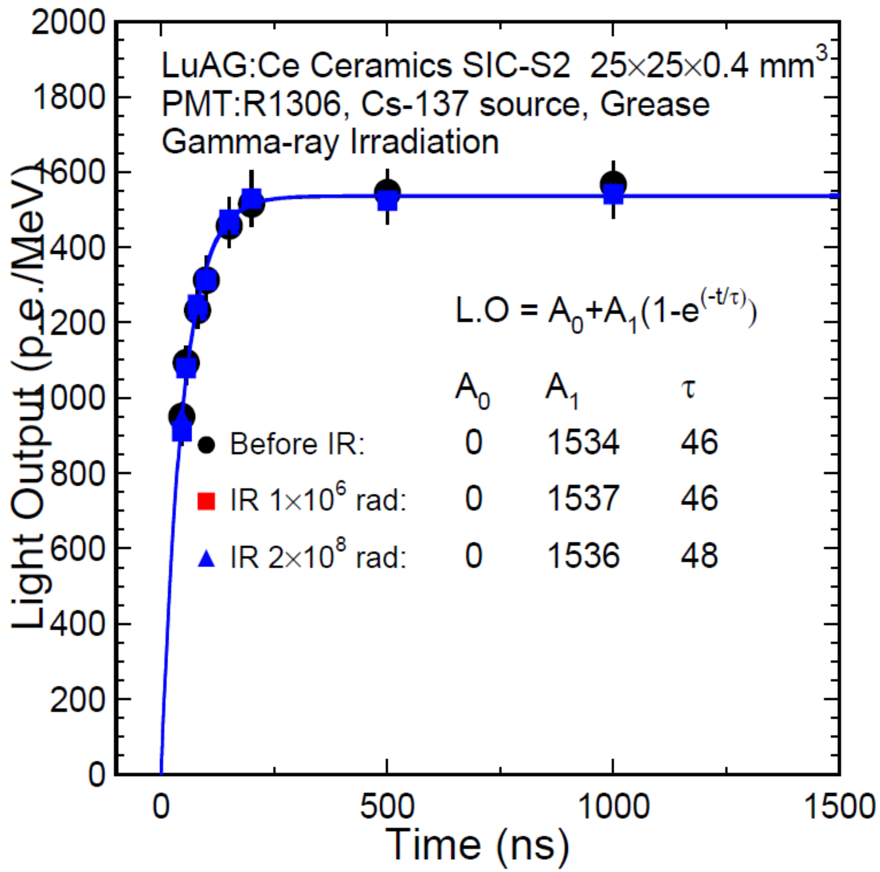
## Experiments

- Properties measured at room temperature: Transmittance, Photo-luminescence, Light Output, Decay Time and Radiation Damage

# Excellent Radiation Hardness

No damage observed in both transmittance and light output after 220 Mrad gamma radiation and $2.9\times10^{14}$ p/cm$^2$ of 800 MeV.

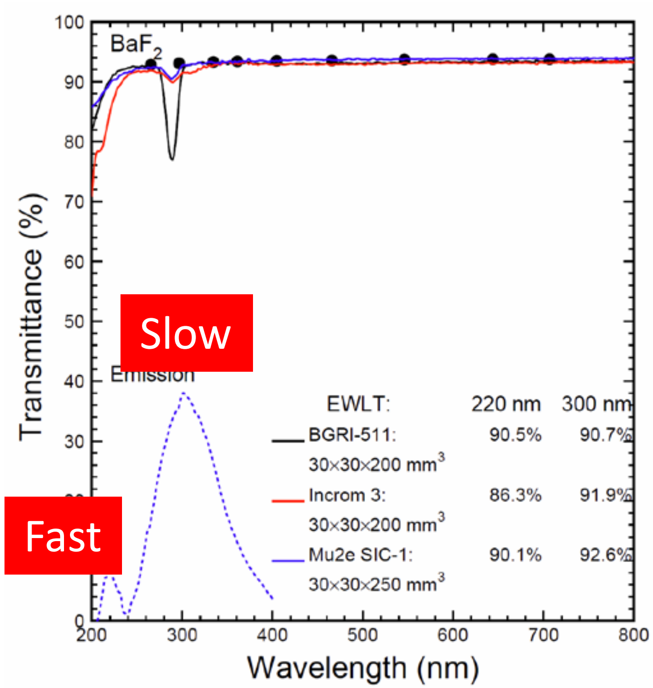


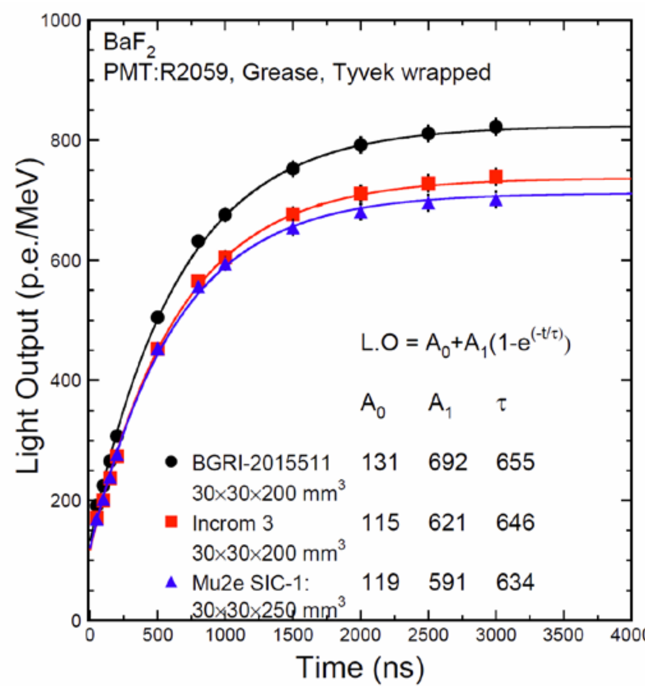




Very promising for a scintillating ceramics based calorimeter

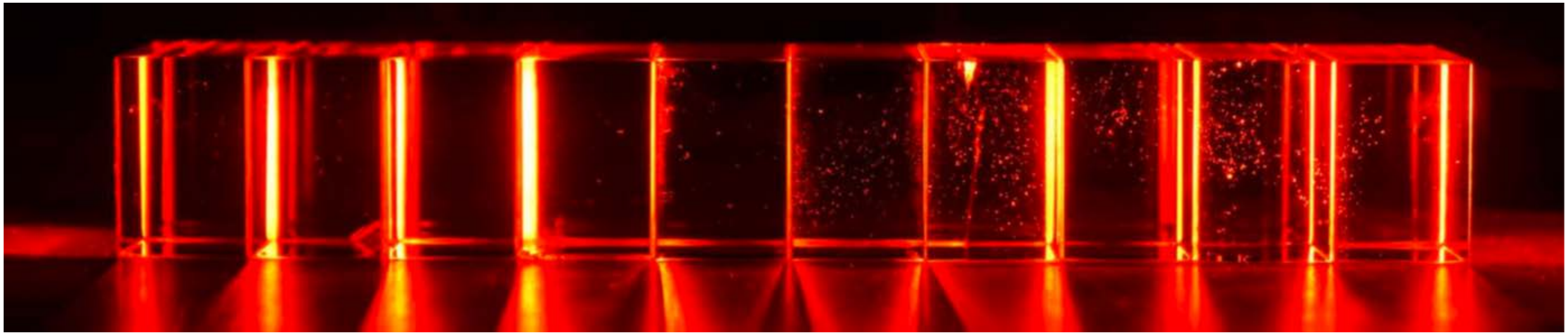
# BaF$_2$: a Promising Scintillator at the Intensity Frontier

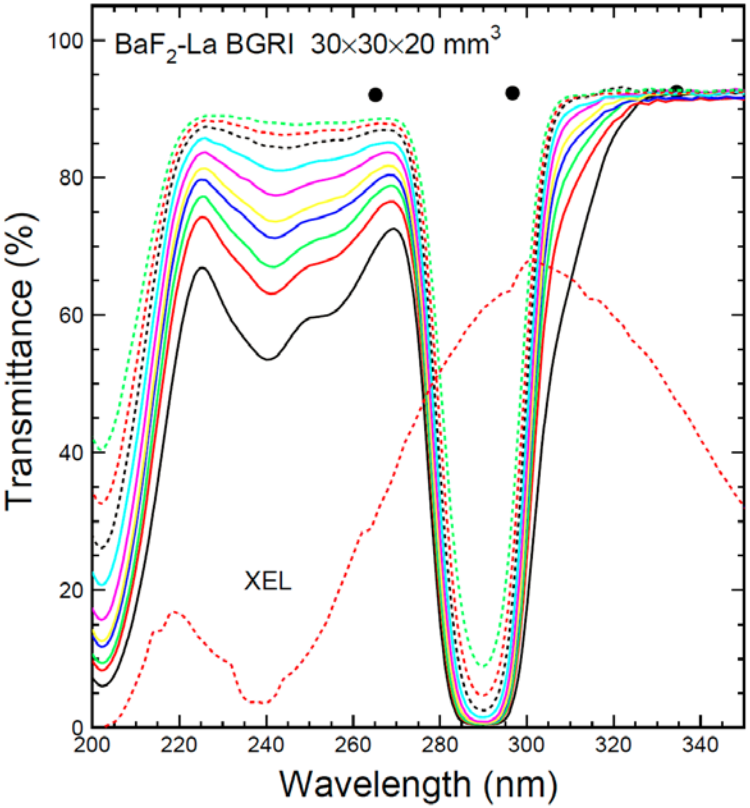
- Good transmittance at both 220 and 300 nm in commercial BaF$_2$ crystals.
- More than 100 p.e./MeV in the fast component, and about a factor of five in the slow component with 600 ns decay time.
- R&Ds with RE doping in BaF$_2$ are ongoing to suppress the slow component.

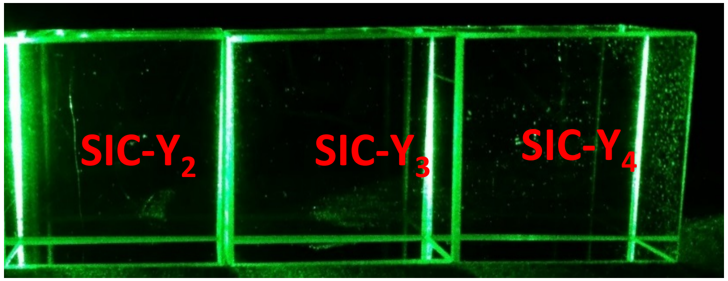
# La/Ce Co-doping at BGRI

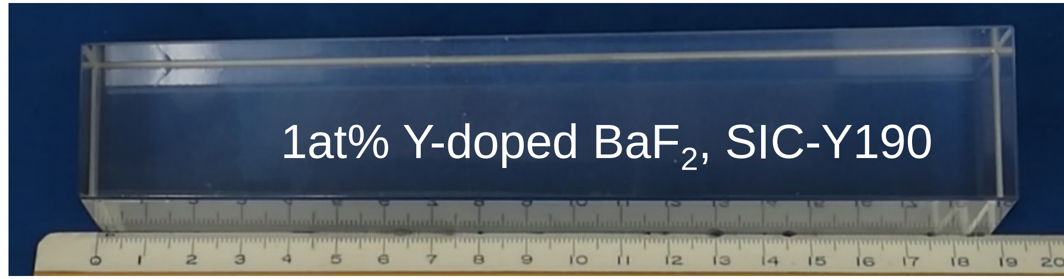


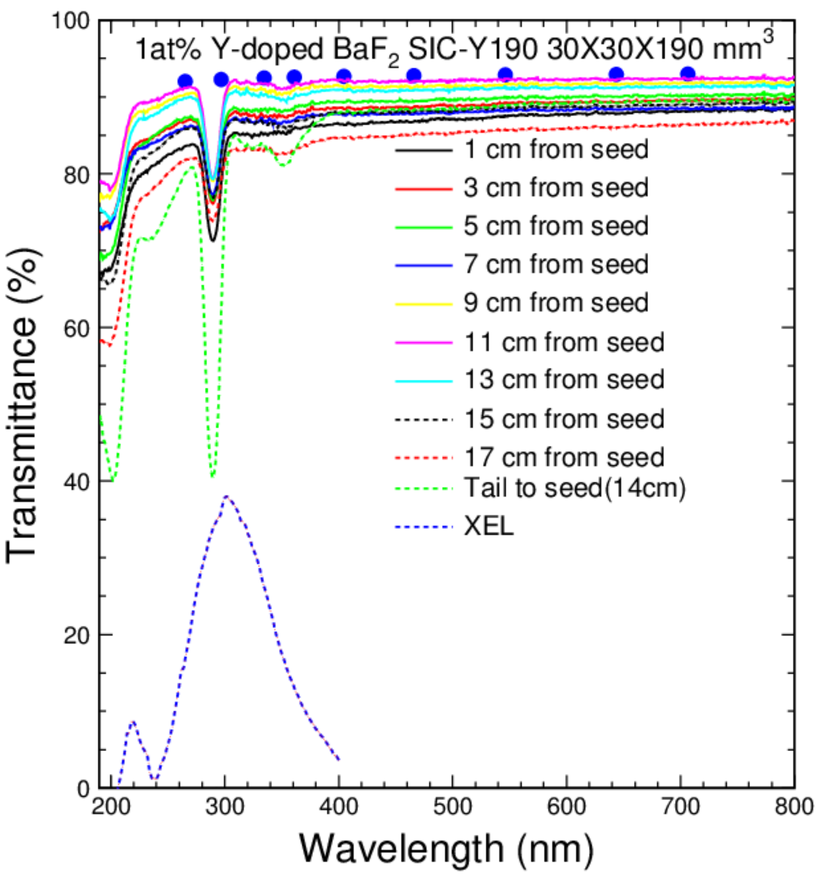



- The La/Ce co-doped sample of BGRI show consistent absorption band of Ce3+ at the slow component.
- The overall F/S ratio is increased from 1:5 to 1:2 in 20 cm long samples with good light response uniformity.
- Their radiation hardness needs further investigation.

# Yttrium doping at SIC



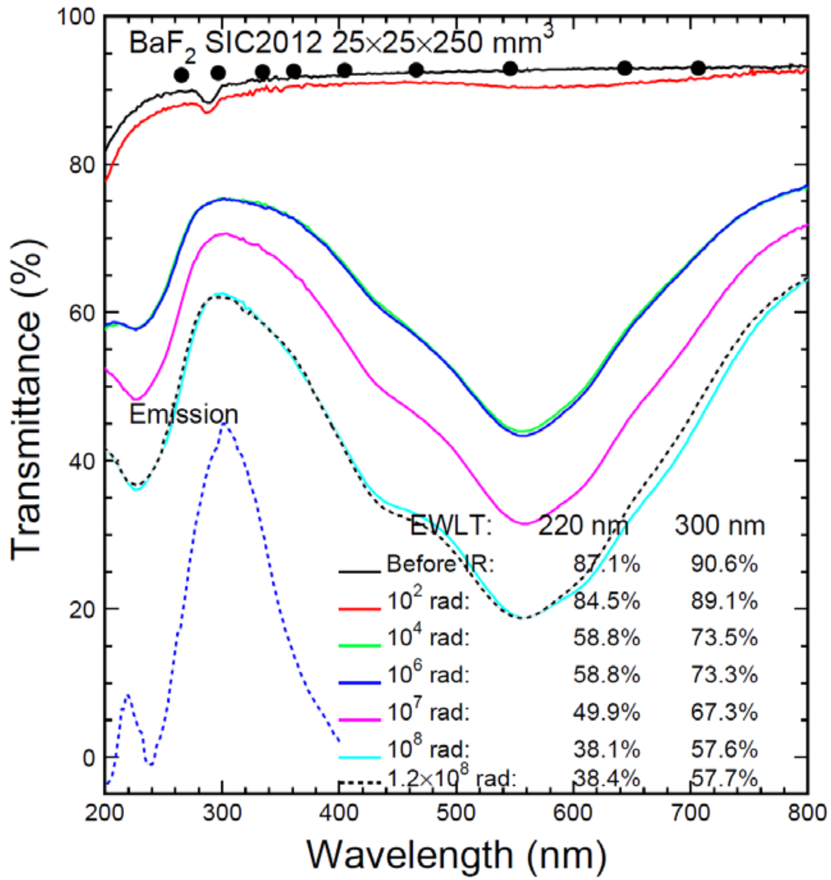



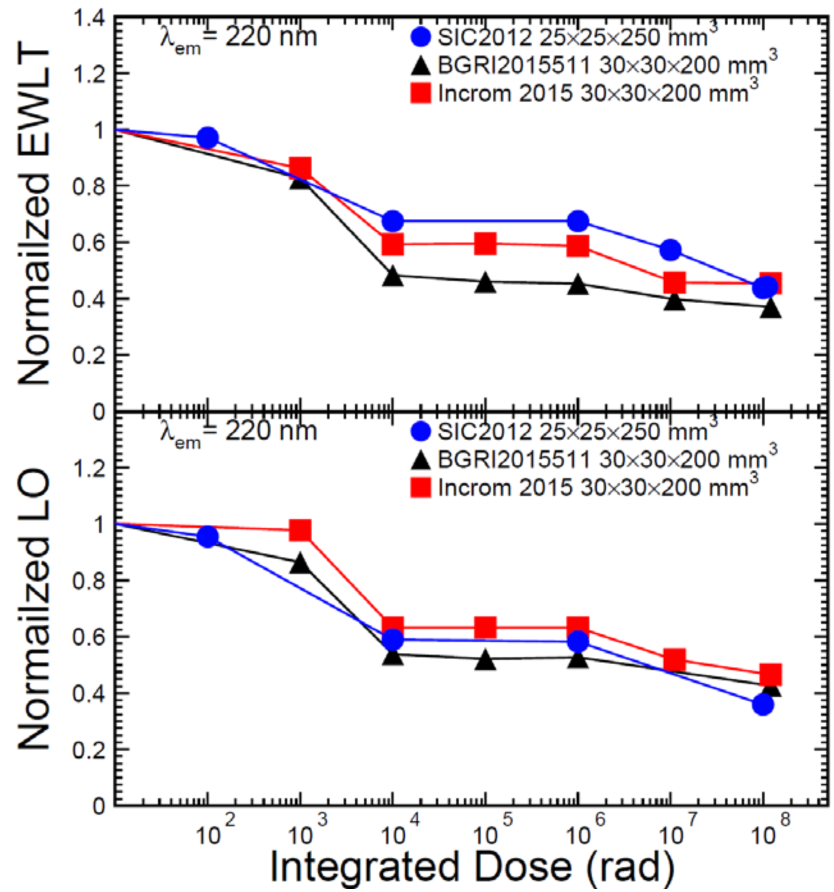




- Y-doped $BaF_2$ crystals of 3 x 3 x 2 $cm^3$ with slow suppression have been successfully grown in SIC and evaluated in Caltech crystal lab.
- F/S ratio of these crystals is increased to 129-146%.
- Y-doped $BaF_2$ crystals up to 20 cm in length have been successfully grown. Its light output, EWLT, F/S ratio and LRU are to be improved.

# Gamma-Ray Induced Damage in $BaF_2$

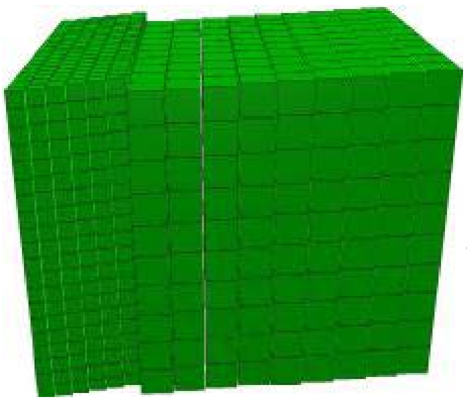
- Consistent γ induced damage in 25 cm $BaF_2$ crystals from three vendors.
- 40%/45% LO of the fast/slow component in 25 cm crystals after 120 Mrad.
- Proton and neutron irradiation tests at LANL are underway.

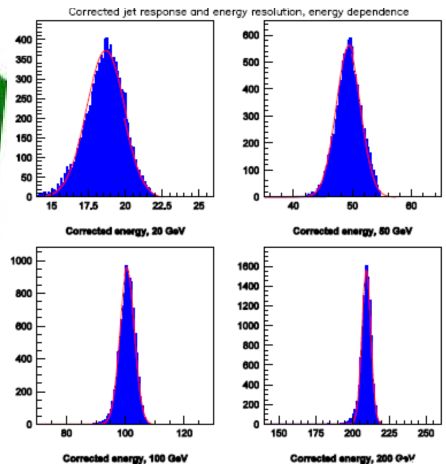


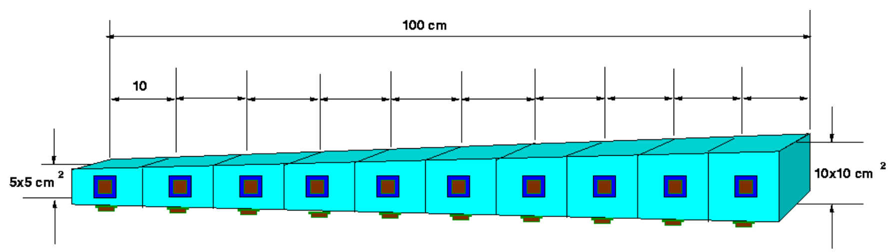



# Homogeneous Hadronic Calorimeter

A Fermilab team (A. Para et al.) proposed a total absorption homogeneous hadronic calorimeter (HHCAL) detector concept to achieve good jet mass resolution by measuring both Cherenkov and Scintillation light.






ILCWS-08, Chicago: a HHCAL cell with pointing geometry

## Requirements for the Materials:

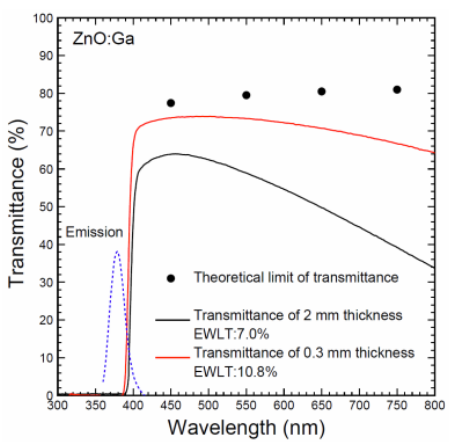
- Cost-effective material: for 70~100 $m^3$
- Short nuclear interaction length: ~ 20 cm.
- Good UV transmittance: UV cut-off < 350 nm, for readout of Cherenkov light.
- Some scintillation light, not necessary bright and fast.
- Discrimination between Cherenkov and scintillation lights, in spectral or temporal domain.

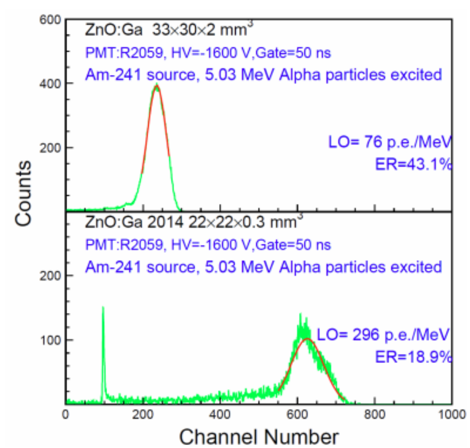
# Candidate Crystals for HHCAL

**Cost-effective, UV transparent crystals with both scintillation and Cherenkov light**

| Parameters | $Bi_4Ge_3O_{12}$ (BGO) | $PbWO_4$ (PWO) | $PbF_2$ | PbClF | $Bi_4Si_3O_{12}$ (BSO) |
|---|---|---|---|---|---|
| ρ (g/cm$^3$) | 7.13 | **8.29** | **7.77** | 7.11 | 6.8 |
| $\lambda_I$ (cm) | 22.8 | **20.7** | **21.0** | 24.3 | 23.1 |
| n @ $\lambda_{max}$ | 2.15 | 2.20 | 1.82 | 2.15 | 2.06 |
| $\tau_{decay}$ (ns) | 300 | 30/10 | **?** | 30 | 100 |
| $\lambda_{max}$ (nm) | 480 | 425/420 | **?** | 420 | 470 |
| Cut-off λ (nm) | **310** | 350 | **250** | **280** | **300** |
| Light Output (%) | 100 | 1.4/0.37 | **?** | 17 | 20 |
| Melting point (°C) | **1050** | **1123** | **842** | **608** | **1030** |
| Raw Material Cost (%) | 100 | 49 | **29** | **29** | 47 |

IEEE Trans. Nucl. Sci. **59** (2012) 2229-2236
IEEE Trans. Nucl. Sci. **61** (2014) 489-494
J. Phys. Conf. Ser. **587** (2015)

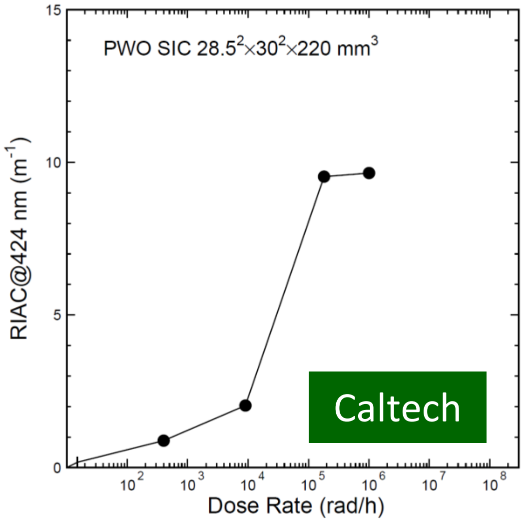

Nano particles made of wide-band semiconductors with self-absorption, such as ZnO, $PbI_2$ , GaAs/InAs etc., may provide alternative composite scintillators featured with tunable emission. Main issues are effective hosts which are transparent for Cherenkov the scintillation light.

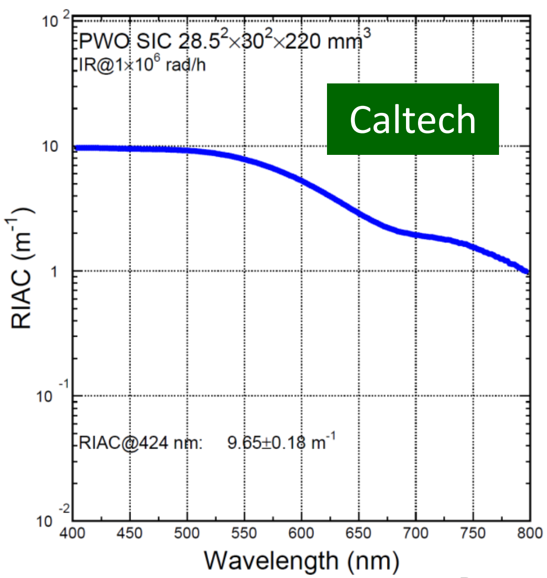
# Summary

- Bright, fast and radiation hard LYSO crystals offer a robust crystal calorimeter for future HEP experiment at the energy frontier. Ongoing is irradiation by protons and neutrons at LANL in 2016.
- Novel inorganic scintillating ceramics, e.g. LuAG:Ce and LuAG:Pr, may offer a cost-effective alternative for LSO/LYSO.
- Fast and radiation hard $BaF_2$ crystals offer a very fast crystal calorimeter for future HEP experiment at the intensity frontier. See David's talk. Slow scintillation suppression by RE doping is underway.
- Cost effective crystals, glasses and ceramics would provide a foundation for a homogeneous hadron calorimeter for future lepton colliders.

# A Comparison of damages in PWO caused by γ-rays and Neutrons up to $10^{19}$ n/cm$^2$

Gamma Irradiation at JPL




7.8×10$^{18}$/1.2×10$^{19}$/4.0×10$^{19}$ n/cm$^2$ for fast/epithermal/thermal
Corresponding dose received: 33 Grad @ 300 Mrad/h

Saclay neutron test:
30 cm$^{-1}$@ 420 nm
under 300 Mrad/h

Caltech gamma test:
0.1 cm$^{-1}$@ 420 nm
Under 1 Mrad/h

Neutron induced damage seems negligible

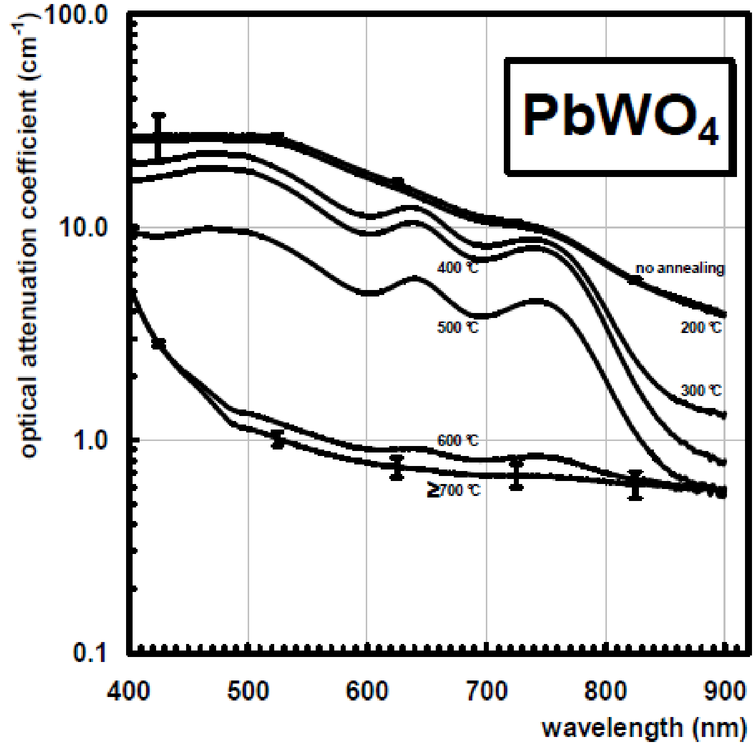



Fig. 2. Optical attenuation coefficient of the irradiated sample before annealing and after successive annealing temperatures.

[50] R. Chipaux et al., ***Behaviour of PWO scintillators after high fluence neutron irradiation,*** in Proc. 8th Int. Conference on Inorganic Scintillators,SCINT2005, A. Getkin and B. Grinyov eds, Alushta, Crimea, Ukraine, September 19–23 (2005), pp. 369–371

# No Neutron Damage in PWO

## 5.2 Radiation damage effects under neutron irradiation

In view of the intense neutron flux expected in CMS (see section 2) the effects on lead tungstate of neutron exposure were studied in nuclear reactors [47, 48]. The neutron fluxes and energies in these exposures were comparable to those expected in CMS. However, in reactors there is a strong associated gamma dose. The effect arising from neutrons was estimated by comparing the reactor results with results obtained from pure gamma irradiations. This indicated that there was no specific effect due to neutrons on the optical and scintillating properties of lead tungstate, at least up to fluences of $10^{14}$ cm$^{-2}$. This was confirmed by later independent studies [49]. It is also to be mentioned that recent tests performed at a very high fluence, of the order of $10^{19}$ to $10^{20}$ n·cm$^{-2}$ and 330 MGy (i.e. well above the level that will be ever achieved in any physics experiment) revealed the robustness of lead tungstate crystals which were not destroyed nor locally vitrified, and remained scintillating after such heavy irradiation [50].

The CMS Electromagnetic Calorimeter Group, ***Radiation hardness qualification of $PbWO_4$ scintillation crystals for the CMS Electromagnetic Calorimeter,*** 2010 JINST 5 P03010