Proton-Induced Radiation Damage in BaF_2, LYSO, and PWO Crystal Scintillators

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***Abstract*— Future high-energy physics experiments at the energy and intensity frontiers will face a challenge of severe radiation environment from both ionization dose and charged and neutral hadrons. The high-luminosity large hadron collider, for example, will present an environment, where up to 130 Mrad ionization dose, 3×10^{14} charged hadrons/cm^2 and 5×10^{15} neutrons/cm^2 are expected. In this paper, we report our investigation on charged hadron-induced radiation damage in BaF_2, LYSO/LFS, and PWO crystals up to 3×10^{15} protons/cm^2 by using 800-MeV protons at the Los Alamos Neutron Science Center. Comparison is made between radiation damages induced by protons and ionization dose alone.**

***Index Terms*— BaF_2, crystals, LYSO, protons, PWO, radiation damage, scintillators.**

I. Introduction

CRYSTAL scintillators are widely used in HEP experiments due to their superb energy resolution and detection efficiency. A lead tungstate ($PbWO_4$ or PWO) crystal calorimeter, for example, has played an important role for the discovery of the Higgs boson by the CMS experiment [1]. One crucial issue, however, is crystal's radiation damage in the severe radiation environment at the large hadron collider (LHC), which requires precision light monitoring to correct variations of crystal's light output (LO) [2]. During the LHC Run I, up to 70% loss of LO was observed by the CMS light monitoring system in the PWO crystals when the LHC was running at a luminosity of 5×10^{33} cm^{-2}s^{-1} and a half of its designed energy [3]. While the dose-rate-dependent damage in PWO crystals are understood to be induced by ionization dose [4], charged hadrons cause cumulative damage [5], [6].

With 5×10^{34} cm^{-2}s^{-1} luminosity and 3000 fb^{-1} integrated luminosity, the high-luminosity LHC (HL-LHC) will present a very severe radiation environment, where up to 130 Mrad ionization dose, 3×10^{14} charged hadrons/cm^2 and 5×10^{15} neutrons/cm^2 will be expected [7]. Bright, fast- and radiation-hard cerium-doped lutetium-yttrium oxyorthosilicate ($Lu_{2(1-x)}Y_{2x}SiO_5$:Ce or LYSO) crystals were proposed to construct an LYSO/W/Quartz capillary sampling calorimeter for the CMS upgrade for the HL-LHC [8]. LYSO crystals were also proposed to construct total absorption calorimeters for the SuperB experiment in Europe [9] and the Mu2e experiment at Fermilab [10]. An LYSO crystal total absorption calorimeter and a 3-D imaging calorimeter are currently being constructed for the COMET experiment at KEK [11] and the HERD experiment in space [12], respectively. LYSO crystals have also been proposed to construct a minimum ionization particle based precision time-of-flight layer for the CMS upgrade for the HL-LHC [13].

Early work indicates that proton-induced radiation damage in LYSO is a factor of five smaller than that in PWO [14]. Experiments carried out by using the 800-MeV protons at the Weapons Neutron Research facility of Los Alamos Neutron Science Center (WNR of LANSCE) and the 24-GeV protons at CERN, however, show much smaller radiation damage [15] and a very good stability of LYSO/W/quartz capillary Shashlik calorimeter cell [16].

The high cost of LYSO, however, limits its use in the future HEP experiments. Other cost-effective fast inorganic scintillators are under investigation. Because of its fast scintillation with subnanosecond decay time, the BaF_2 is being considered as one candidate for Mu2e upgrade at Fermilab [17].

Proton-induced radiation damage was investigated for LYSO, BaF_2, and PWO [14], [15], [18]–[22]. In these studies, proton-induced ionization dose was calculated by using *dE/dx*. It was concluded that the ionization dose induced by protons could not explain the observed damage in crystals. The results of the above investigations as well as this investigation, thus, are presented as a function of the proton fluence. Because of the radioactivity induced by protons in the crystals, most of these measurements were carried out several tens of days after irradiation. In this paper, we report proton-induced radiation damage measured *in situ* before and immediately after irradiation by the 800-MeV proton beam at LANSCE for LYSO, BaF_2, and PWO crystals. Optical and scintillation properties of these samples were also characterized at Caltech before irradiation and after crystals samples are cooled down. In addition, three batches of thin crystal plates of LYSO, BaF_2, and PWO irradiated to various proton fluence were also measured at Caltech, and are compare to the online data.

II. Experimental Setup and Samples

The experiments 6501, 6990, and 7324 were carried out in 2014, 2015, and 2016, respectively, by using 800-MeV

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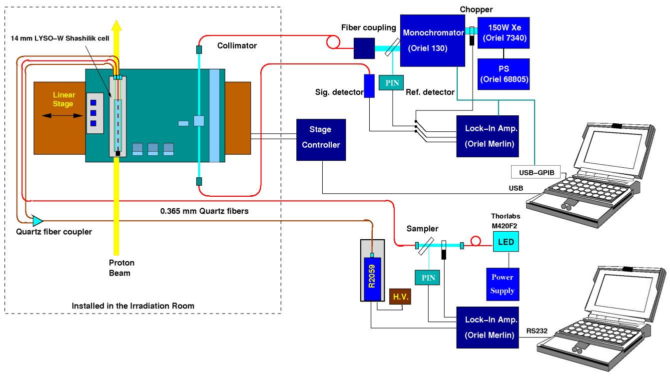

Fig. 1. Schematic showing the experimental setup used to measure crystal's transmittance spectra and to monitor the response for a Shashlik cell *in situ* during the experiment 7324 in 2016.

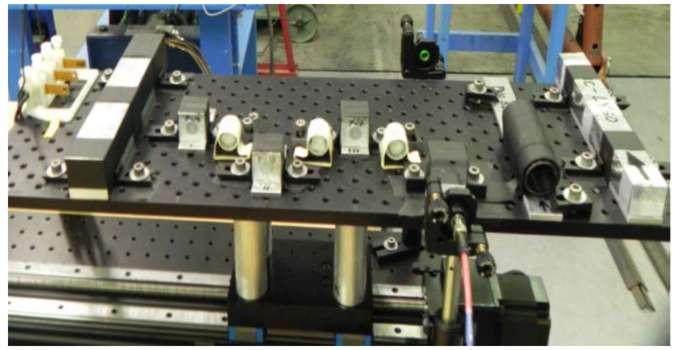

Fig. 2. Photograph showing the eight samples on the linear stage in the experiment 7324 in 2016.

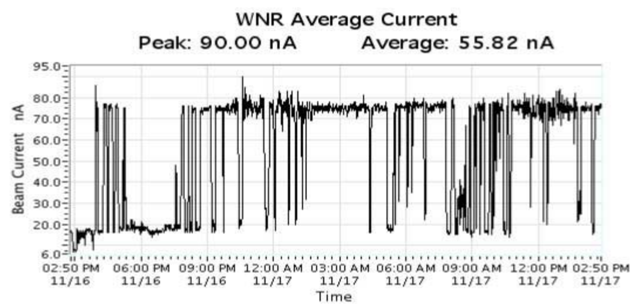

Fig. 3. Proton beam history of the experiment 7324 in 2016.

protons in the blue room of LANSCE. The proton beam had a Gaussian shape with a full-width at half-maximum of about 2.5 cm.

Fig. 1 is a schematic showing the setup, consisting of a linear translation stage, an optical fiber, and lock-in amplifier-based spectrophotometer used to measure crystal's transmittance, and an LED-based monitoring system used to measure response of an LYSO/W/Quartz capillary Shashlik cell *in situ* at LANSCE. The linear stage with a travel distance of 1 m was used to move each sample into the proton beam via a remote control. A part of the chopped light from a 150 W Xe lamp through a monochromator was monitored by a reference photodiode (Thorlabs DET10A). The main part of the light was injected into the crystal sample via Φ 0.365-mm quartz fibers and through two collimators at the front and back of the crystal, and was measured by a signal photodiode (Oriel 70336). The lock-in amplifier (Oriel Merlin) measured the ratio between the signal and reference photodetectors. The precision and stability of this ratio is about 1%, and is free from both the fluctuations of the light intensity of the lamp- and the radiation-induced phosphorescence background in the sample. The LED-based monitoring system was used to measure the response of lutetium fine silicate (LFS from Zecotek Inc., with performances similar to LYSO)/W/Quartz capillary Shashlik cells *in situ* before, during, and immediately after irradiation, in which a lock-in amplifier was also used to mitigate radiation-induced phosphorescence.

Fig. 2 shows the samples loaded on the stage during the experiment 7324 in 2016. Because of the large distance between the samples, multiple scattering effects to the neighboring crystal were negligible. Fig. 3 shows the history of the corresponding proton beam current, which was used to calculate the fluence applied to each sample during the experiment.

Table I lists the sample dimension and the proton fluence. In addition to online measurements, the longitudinal transmittance (LT) and LO were also measured at Caltech for all samples before and after the experiment. Transmittance was measured by a PerkinElmer Lambda 950 spectrophotometer with 0.15% precision. The precision for the corresponding radiation-induced absorption coefficient (RIAC) is about 3.5 m^{-1} for 1.5-mm-thick LYSO plates, and is much less for long crystals. To reduce the systematic uncertainties caused by variations of the beam position at a few millimeters during irradiation all the transmittance measured at Caltech after irradiation is an average of the transmittance measured at four points symmetrically distributed in the front face of the crystals.

The LO of LYSO thin plates before and after irradiation was measured by using 0.662-MeV γ-rays from a ^{137}Cs source with grease coupled to a Hamamatsu R1306 PMT. The LO of BaF_2 and PWO thin plates before and after irradiation was measured by using 0.511-MeV γ-rays from a ^{22}Na source with grease coupled to a Hamamatsu R2059 PMT. The systematic uncertainty of the LO measurements is about 1%.

III. Experimental Results

A. BaF_2 Crystals

Fig. 4 shows the LT spectra (330–500 nm) measured *in situ* for a 20-mm BaF_2 sample from Hellma Materials GmbH before and after irradiation from 6.1×10^{12} p/cm^2 to 1.2×10^{15} p/cm^2 in six steps, as well as at 9 h after the irradiation. The online spectrophotometer was cross calibrated by using the LT spectrum data measured by the PerkinElmer LAMBDA 950 spectrophotometer at Caltech. The results indicate that this BaF_2 crystal is transparent in the deep ultraviolet region where its fast emissions are peaked. The observed recovery can be attributed to a thermal relaxation of the crystal.

Fig. 5 shows the LT spectra between 200 and 500 nm measured at Caltech for three 25 × 25 × 5 mm^3 BaF_2 plates from the Shanghai Institute of Ceramics (SIC) after 2.7, 16, and 97 × 10^{13} p/cm^2 irradiation. The blue dashed

TABLE I
SAMPLES AND 800-MEV PROTON FLUENCE

No.	Samples	Dimensions (mm^3)	Fluence (p/cm^2)	Experiment
1	BaF_2	30×30×20	6.1×10^{12} – 1.2×10^{15}	7324
2	BaF_2	30×30×20	2.9×10^{14}	6990
3	BGO	25×25×200	1.8×10^{14}	6990
4	BGO	17×17×17	2.9×10^{14}	6990
5	SIC LYSO	25×25×200	5.0×10^{13} - 3.0×10^{15}	7324
6	LFS	25×25×180	1.8×10^{14} – 2.9×10^{15}	6990
7	SG LYSO	25×25×200	1.6×10^{14} - 3.3×10^{14}	6501
8	Shashlik (LFS/W/Capillary)	34×34×215	1.2×10^{15} – 1.9×10^{15}	6990 and 7324
9	PWO	25×25×5	4.4×10^{12} – 1.2×10^{15}	7324
10	PWO	$28.5^2\times30^2\times220$	1.8×10^{14}	6990
11	2×PWOs, 2×BaF_2	25×25×5	2.7×10^{13}	7324
	3×LYSO	10×10×3		
12	2×PWOs, 2×BaF_2	25×25×5	1.6×10^{14}	7324
	3×LYSO	10×10×3		
13	2×PWOs, 2×BaF_2	25×25×5	9.7×10^{14}	7324
	3×LYSO	10×10×3		

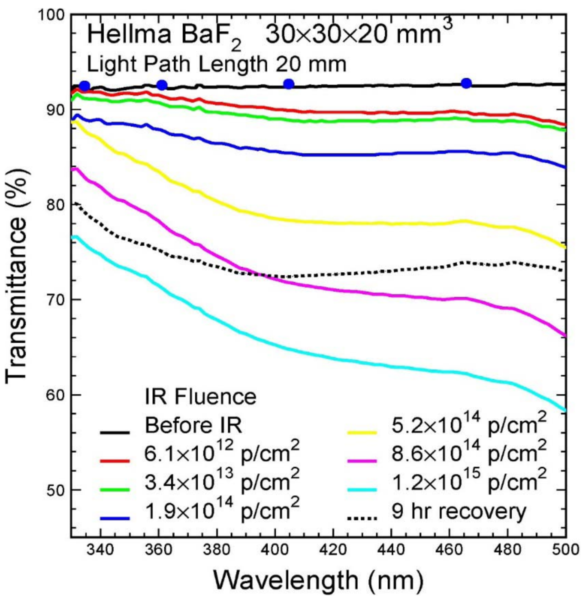

Fig. 4. LT spectra of a BaF_2 crystal before and after irradiations.

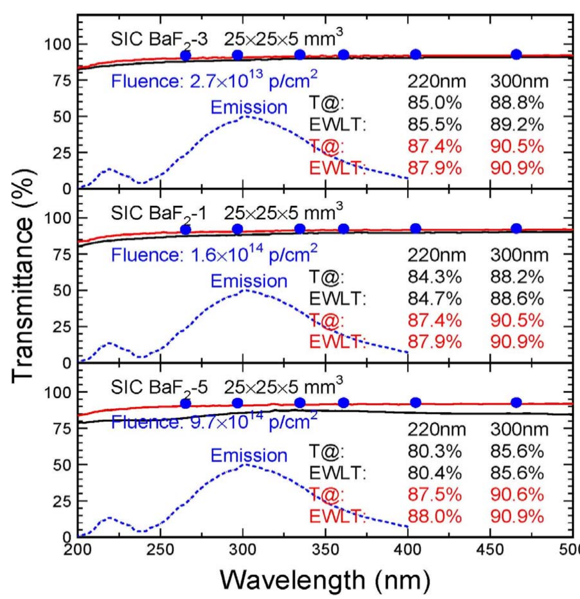

Fig. 5. LT spectra of three BaF_2 samples before and after irradiations.

line in Fig. 5 shows the emission spectra of BaF_2 with two emission peaks at 220 and 300 nm, respectively, for a fast component with subnanosecond decay time and a slow component with 600-ns decay time. Also listed in Fig. 5 are the numerical values of transmittance at these two emission peaks and the emission-weighted LT (EWLT) [23]. The EWLT value provides a numerical representation of the LT data across the emission spectrum, so is a direct measure of transparency for crystal's scintillation light. The EWLT values at 220 nm are 85.5%, 84.7%, and 80.4% after 2.7, 16, and 97 × 10^{13} p/cm^2 irradiation. The 9% loss of the BaF_2 plate's transmittance at the fast scintillation peak of 220 nm after 9.7 × 10^{14} p/cm^2 indicates its excellent radiation hardness against proton.

Fig. 6 shows the LO as a function of integration time and corresponding exponential fit for three 5-mm BaF_2 plates after 2.7, 16, and 97 × 10^{13} p/cm^2. Also listed in Fig. 6 are the LO integrated in 50 and 2500 ns, as well as the numerical values of the LO for the fast and slow component obtained from the fit. The 50-ns LO values are 310, 314, and 310 p.e./MeV, respectively, after 2.7, 16, and 97 × 10^{13} p/cm^2 for three BaF_2 samples. The average loss of 50-ns LO is at about 9% after 9.7 × 10^{14} p/cm^2, confirming its excellent radiation hardness against protons.

B. PWO Crystals

Fig. 7 shows the LT spectra measured *in situ* for an SIC PWO sample of 25 × 25 × 5 mm^3 before and after irradiation from 1.6 × 10^{13} p/cm^2 to 1.2 × 10^{15} p/cm^2 in six steps as well as 10 h after the irradiation. Also shown in Fig. 7 are the PWO emission in blue dashed lines and the numerical values

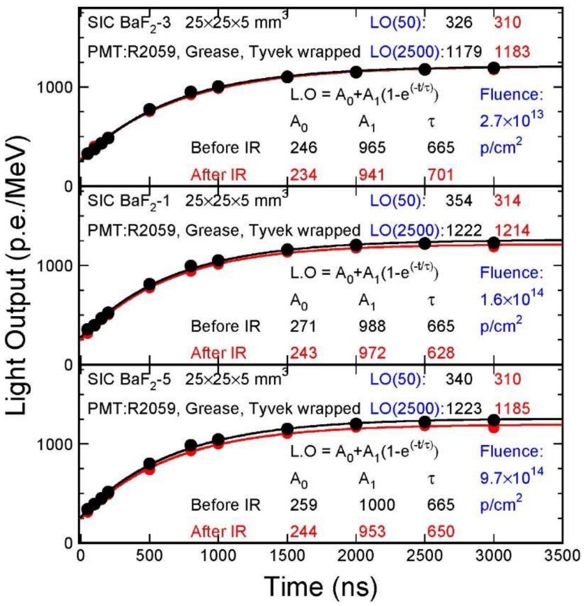

Fig. 6. LO and decay of three BaF_2 crystals before and after irradiation.

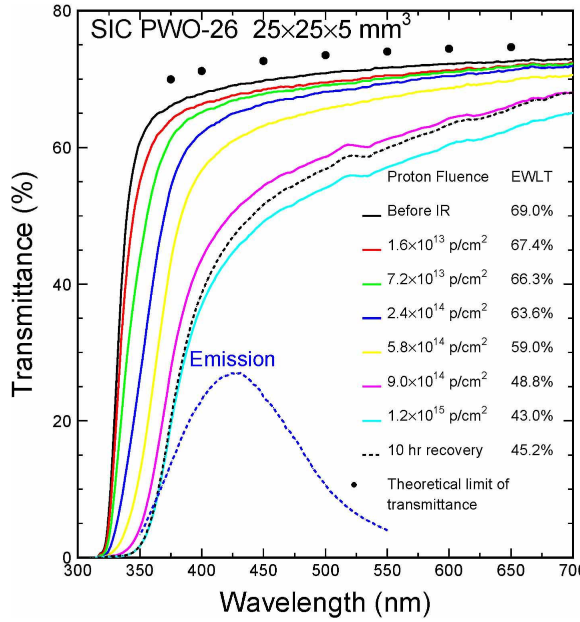

Fig. 7. LT spectra of a PWO crystal before and after irradiations.

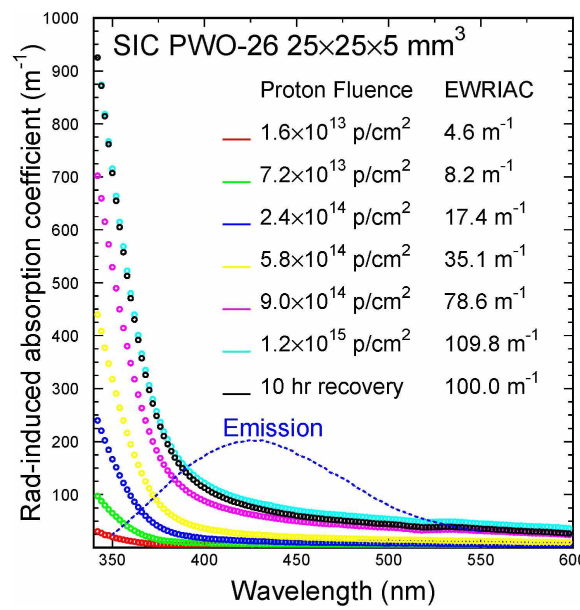

Fig. 8. RIAC spectra of a PWO sample before and after the irradiations.

of EWLT. The EWLT values are 63.6%, 43%, and 45.2 % after 2.4×10^{14}, 1.2×10^{15} p/cm^2, and 10-h recovery. The EWLT loss is over 35% after proton irradiation of 1.2×10^{15} p/cm^2, which is much higher than that of BaF_2 plates with the same dimensions and similar proton fluence, as shown in Fig. 5. The corresponding RIAC [25], [26] spectra are shown in Fig. 8, where the numerical values of the emission-weighted RIAC (EWRIAC) are also listed. The RIAC values at its emission peak 430 nm are 11.8, 83.2, and 73.3 m^{-1}, respectively, after 2.4×10^{14}, 1.2×10^{15} p/cm^2 irradiation, and 10-h recovery, with corresponding EWRIAC values of 17.4, 109.8, and 100 m^{-1}, respectively, indicating severe radiation damage caused by protons in PWO crystal. This result is consistent with our previous report for a 22-cm-long PWO sample irradiated at LANSCE in the experiment 6990 in 2015 [25], where LT below 450 nm was not measurable after 1.8×10^{14} p/cm^2 because of the very large values of RIAC and the long light path length.

C. LYSO Crystals

Fig. 9 shows the LT spectra measured *in situ* for a 20-cm-long LYSO from SIC before and after irradiation. Also listed in Fig. 9 are the LYSO emission in blue dashed lines and the numerical values of EWLT. The EWLT values are 49.6%, 32.9%, 10.5%, and 13.6% after 5×10^{13}, 4×10^{14}, 3×10^{15} p/cm^2, and 24-h recovery. The corresponding RIAC spectra are shown in Fig. 10. The RIAC values at 430 nm are 2.6, 9.6, and 8.1 m^{-1}, respectively, after 4×10^{14}, 3×10^{15} p/cm^2, and 24-h recovery, which are consistent with our previous results [15], [25]. The EWRIAC value of 11.6 m^{-1} after 3×10^{15} p/cm^2 proton irradiation is a factor of 10 smaller than that of PWO.

An LYSO/W/Quartz capillary Shashlik cell consisting of 30 LFS plates of $14 \times 14 \times 1.5$ mm^3 and 29 tungsten plates of $14 \times 14 \times 2.5$ mm^3 interleaved with 15-μm-thick Al foils was constructed [8], [24], and was tested in the experiment 6990 in 2015. Fig. 11 shows the monitoring response as a function of proton fluence. The LO losses of 10% and 50% after 3×10^{14} and 1.2×10^{15} p/cm^2, respectively, confirm LYSO's excellent radiation hardness against charged hadrons. Research and Development work will continue to further develop this inorganic crystal scintillator-based detector concept to provide a fast, radiation hard and cost-effective calorimeter for future HEP experiments at the energy frontier.

D. Comparisons and Correlations

Fig. 12 summarizes the EWRIAC values as a function of the proton fluence for BaF_2, LYSO, and PWO crystals. They are

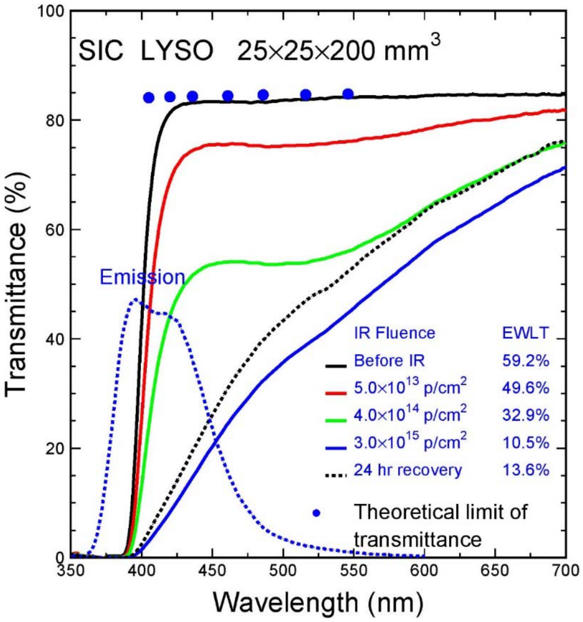

Fig. 9. LT spectra of an LYSO crystal before and after the irradiations.

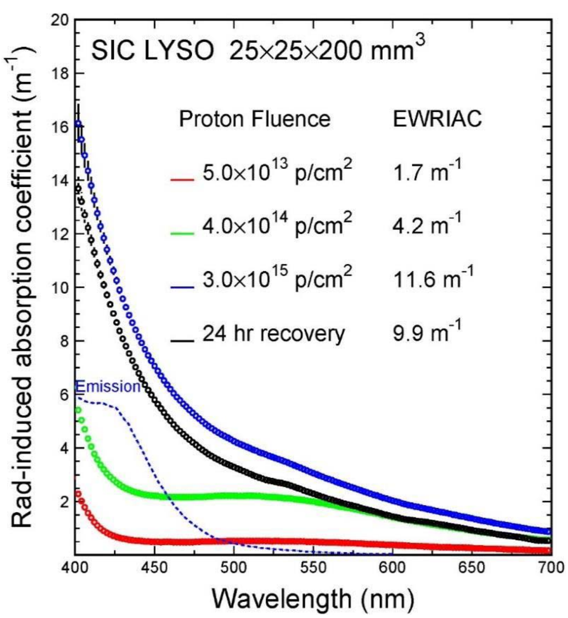

Fig. 10. RIAC spectra are shown for an LYSO sample before and after the irradiation.

calculated by using the LT spectra data measured before and 181 days after the irradiations by the PerkinElmer LAMBDA 950 spectrophotometer at Caltech. They are 7, 18, and 71 m^{-1}, respectively, for LYSO, BaF_2, and PWO after a proton fluence of 9.7×10^{14} p/cm^2, indicating excellent radiation hardness of LYSO and BaF_2 crystals against charged hadrons.

Fig. 13 summarizes the normalized LO as a function of the proton fluence for BaF_2, LYSO, and PWO, measured before and after the proton irradiation at Caltech. The LO losses are 10% and 13%, respectively, for the LYSO and BaF_2 samples after a proton fluence of 9.7×10^{14} p/cm^2, confirming the excellent radiation hardness of LYSO and BaF_2 crystals against charged hadrons. The LO of PWO samples after proton

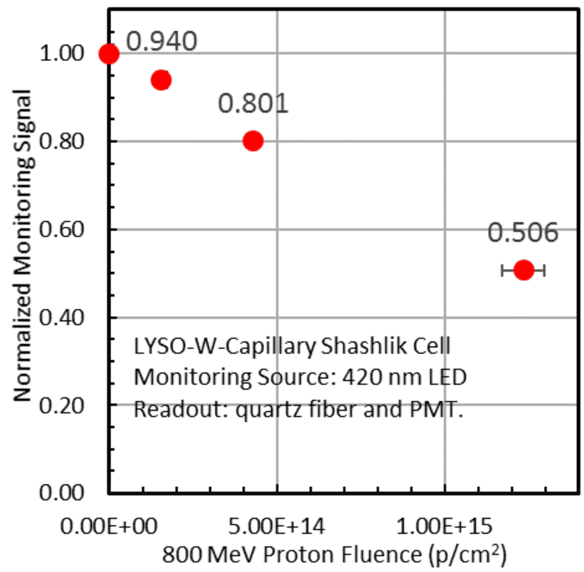

Fig. 11. Monitoring response of an LYSO/Quartz capillary Shashlik cell is shown as a function of the proton fluence.

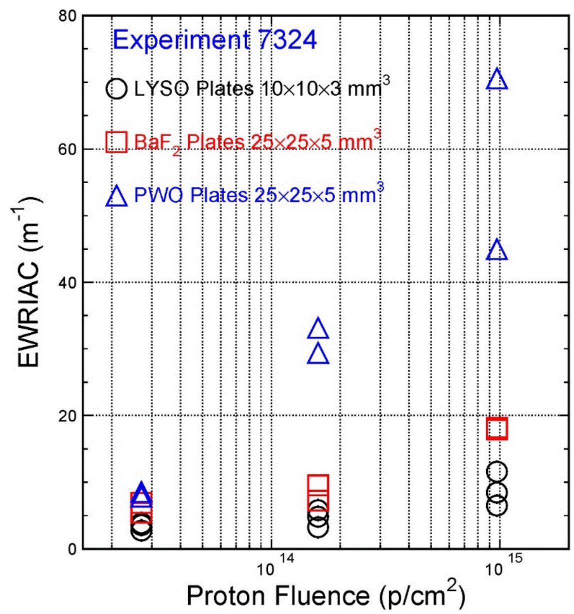

Fig. 12. EWRIAC values are shown as a function of the proton fluence for BaF_2, LYSO, and PWO crystals.

fluence of 9.7×10^{14} p/cm^2 is too low to be experimentally determined.

Fig. 14 shows the normalized LO with an integration gate of 50 ns as a function of EWRIAC of the 220-nm peak for the SIC BaF_2 samples under various irradiation conditions. The blue dots represent the results for proton irradiation, while the red square represents the neutron plus gamma [26] irradiation results. The EWRIAC value of 18 m^{-1} after 9.7×10^{14} p/cm^2 proton irradiation causes an LO loss of about 13% for $25 \times 25 \times 5$ mm^3 plates. The exponential fit for the normalized

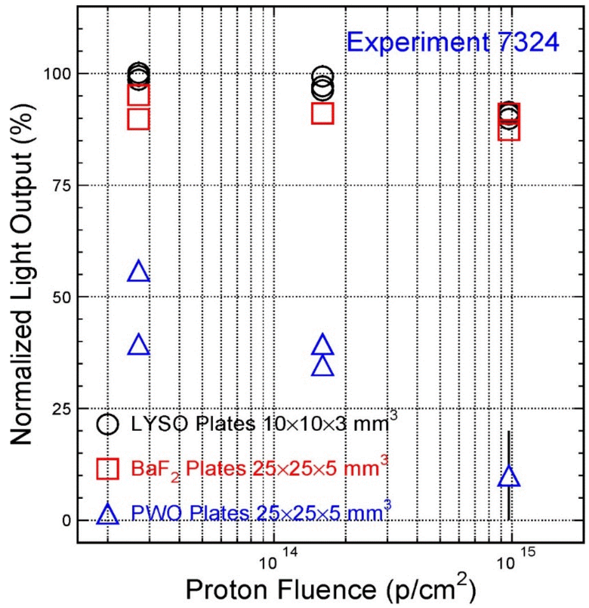

Fig. 13. Normalized LO is shown as a function of the proton fluence for BaF_2, LYSO, and PWO crystals.

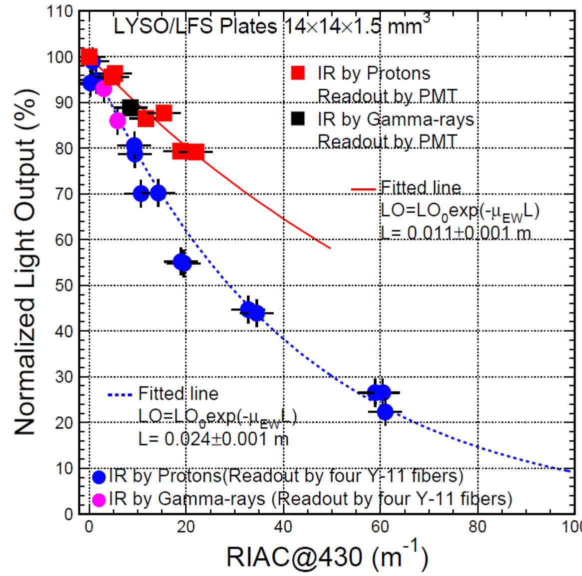

Fig. 15. Normalized LO is shown as a function of the RIAC values at 430 nm for LYSO/LFS crystals.

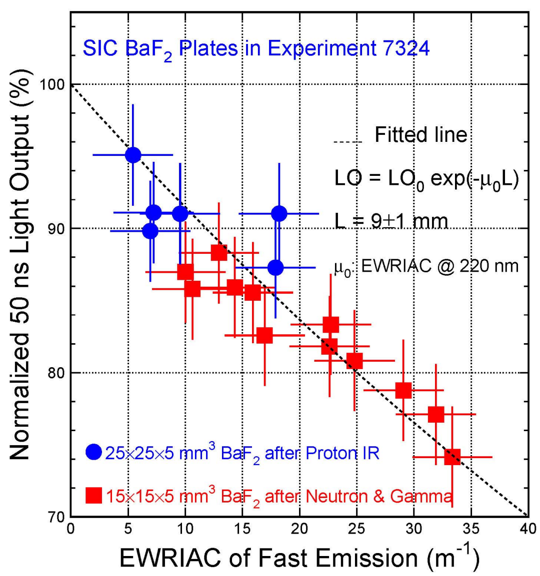

Fig. 14. Normalized LO in 50 ns is shown as a function of the EWRIAC values for BaF_2 crystals.

LO versus the EWRIAC values indicates an average of 9-mm optical path length of the BaF_2 scintillation light in both the $25 \times 25 \times$ 5mm^3 and $15 \times 15 \times 5$ mm^3 plates. The consistent correlation between the normalized LO loss and EWRIAC for damages induced by protons and neutron/γ-rays with a 9-mm light path length indicates that the LO loss can be monitored by measuring crystal's transparency independent of the radiation source. It is thus clear that the LO loss in BaF_2 crystals can be corrected by using a precision monitoring system in calorimeter application. A detailed comparison between the radiation damage caused by ionization dose and charged hadrons will be discussed in our future publications.

Fig. 15 shows the normalized LO loss as a function of the RIAC values at 430 nm for $14 \times 14 \times 1.5$ mm^3 LYSO plates. The LO of these plates was measured by two methods: 1) through four Y-11 wavelength shifting fibers coupled to a PMT or 2) directly coupling the LYSO plates to the PMT [8]. The data comes from the LYSO/LFS plates irradiated by proton (red and blue) and γ-rays (black and pink) [27]. The correlations between the normalized LO and RIAC caused by protons and γ-rays are consistent with average light path length of 2.4 and 1.1 cm, respectively, obtained by an exponential fit for these two coupling methods. Once again, this consistency between the normalized LO loss and RIAC for damages induced by protons and neutron/γ-rays indicates that the LO loss can be monitored by measuring crystal's transparency independent of the radiation source.

IV. Summary

LYSO/LFS crystals show consistent EWRIAC of $\sim$3 m^{-1} after 3×10^{14} p/cm^2 by either 800-MeV or 24-GeV protons, indicating excellent radiation hardness. An LO loss at a level of 10% was observed in an LYSO/quartz capillary shashlik cell after 3×10^{14} p/cm^2 by 800-MeV protons at LANSCE, indicating a robust calorimeter.

The EWRIAC value and the LO loss are about 20 m^{-1} and 10%, respectively, after 10^{15} p/cm^2 in BaF_2 crystals, indicating excellent radiation hardness.

Consistent relations between the LO loss and radiation-induced absorption are observed between damages caused by ionization dose, protons, and neutrons in both LYSO and BaF_2 crystals, indicating that the LO loss in these crystals can be monitored by measuring crystal's transparency independent of the nature of the radiation sources.

In a brief summary, LYSO and BaF_2 crystals survive the severe radiation environment expected at the HL-LHC, so will serve as excellent candidate crystal detectors for future HEP experiments at the HL-LHC. Also reported in this conference are the results of investigations on radiation damage induced by neutron [26]. While these results are crucial for crystal's applications in the future HEP experiments, further investigations are still needed to fully understand the nature of radiation damages caused by hadrons, including both charged and neutral hadrons.

The result of this investigation also provides an important information for understanding proton-induced radiation damage in these crystals, which will be an important input for their use in the future HEP experiments at both the energy and intensity frontiers.

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