

## Study of radiation hardness of $\text{Gd}_2\text{SiO}_5$ scintillator for heavy ion beam

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## TECHNICAL REPORT

## Study of radiation hardness of $\text{Gd}_2\text{SiO}_5$ scintillator for heavy ion beam

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**ABSTRACT:**  $\text{Gd}_2\text{SiO}_5$  (GSO) scintillator has very excellent radiation resistance, a fast decay time and a large light yield. Because of these features, GSO scintillator is a suitable material for high radiation environment experiments such as those encountered at high energy accelerators. The radiation hardness of GSO has been measured with Carbon ion beams at the Heavy Ion Medical Accelerator in Chiba (HIMAC). During two nights of irradiation the GSO received a total radiation dose of  $7 \times 10^5$  Gy and no decrease of light yield was observed. On the other hand an increase of light yield by 25% was observed. The increase is proportional to the total dose, increasing at a rate of 0.025%/Gy and saturating at around 1 kGy. Recovery to the initial light yield was also observed during the day between two nights of radiation exposure. The recovery was observed to have a slow exponential time constant of approximately  $1.5 \times 10^4$  seconds together with a faster component. In case of the LHCf experiment, a very forward region experiment on LHC (pseudorapidity  $\eta > 8.4$ ), the irradiation dose is expected to be approximately 100 Gy for  $10 \text{ nb}^{-1}$  of data taking at  $\sqrt{s} = 14 \text{ TeV}$ . The expected increase in light yield of less than a few percent will not affect the LHCf measurement.

**KEYWORDS:** Radiation-hard detectors; Scintillators, scintillation and light emission processes (solid, gas and liquid scintillators); Accelerator Applications; Calorimeters

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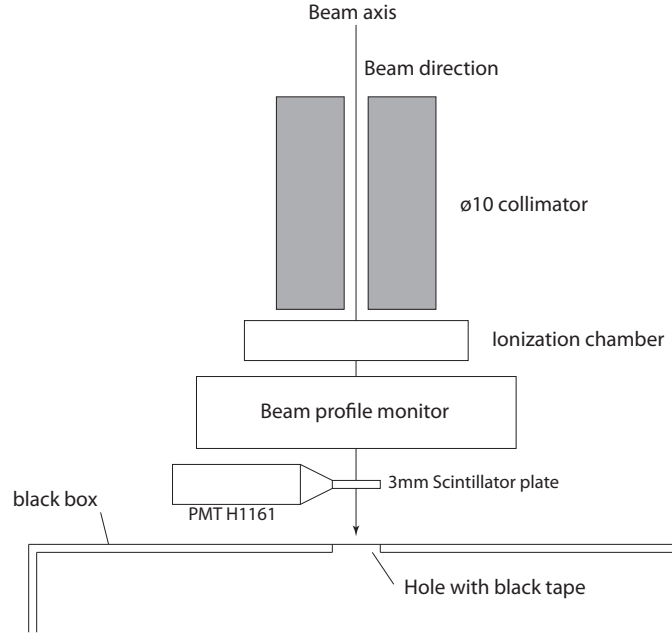
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## 1 Introduction

The radiation resistance of particle detectors is an important issue for the latest very high intensity accelerator experiments. In case of some scintillators, irradiation results in a significant decrease in light yield and transmittance [1] on the scale of irradiation expected and these must be avoided. Materials having good radiation hardness are required for such experiments.

Cerium-doped GSO scintillator ( $\text{Gd}_2\text{SiO}_5:\text{Ce}$ ) is known to have a very strong radiation resistance, a fast decay time (30 to 60ns) among inorganic scintillators, and a large amount of light yield (20% of NaI) [2]. The properties of the GSO scintillator have been investigated in previous studies [2–6]. By using a  $^{60}\text{Co}$  gamma-ray source, no significant decrease in transmittance was observed up to  $10^7$  Gy in the measurement of [3], however an increase in light yield was observed in the measurement of [2]. On the other hand, a small degradation of transmission of GSO scintillator for  $^{60}\text{Co}$  gamma-ray dose of  $10^5$  Gy was observed in the measurement of [4]. In a proton exposure experiment, no significant degradation of transmittance up to  $10^4$  Gy and sizable degradation at  $10^5$  Gy were reported [5]. It seems that the radiation hardness against proton is by two orders of magnitude smaller compared with gamma-ray irradiation. In a measurement under 50MeV  $^{12}\text{C}$  irradiation up to  $10^7$  Gy, large and small decreases in light yield and transmittance, respectively, were reported [6]. In the study reported in this paper, a complementary measurement was performed by using high energy Carbon (290MeV/n) ion beams at the Heavy Ion Medical Accelerator in Chiba (HIMAC). The light yield of GSO scintillator under irradiation was investigated and the radiation hardness up to  $10^6$  Gy was measured. In section 2, the experimental setup is described. In section 3, the experimental data and discussions of the results and their impact on LHCf measurement [1] are presented. The motivation for the work reported in this paper is the consideration of a possible upgrade of the LHCf detectors as the LHC collision energy is increased from  $\sqrt{s} = 7$  TeV to  $\sqrt{s} = 14$  TeV.



**Figure 1.** The experimental setup along the beam axis.

## 2 Experimental setup

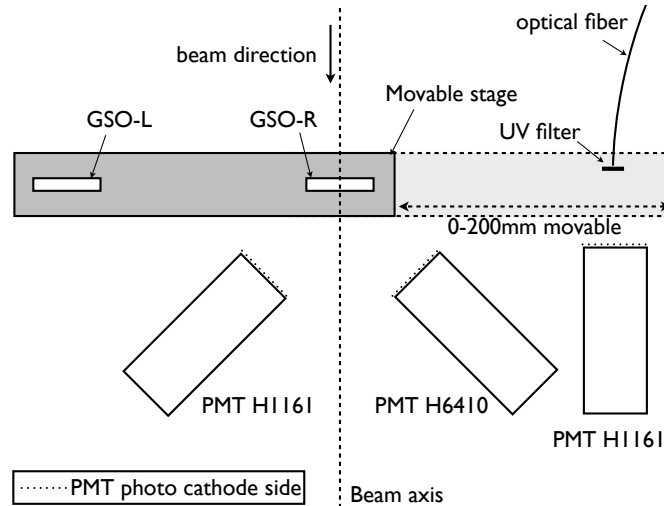
The radiation hardness of GSO scintillator was examined by using a Carbon ( $^{12}\text{C}$ ) ion beam with energy of 290 MeV/n (Total 3.48 GeV/ion). Irradiation was carried out over two nights (9 and 10 Nov 2010) at HIMAC. During the first night, the irradiation rate increased in steps from  $\sim 10^7$  to  $\sim 4 \times 10^9$  particles per spill. For the second night, the irradiation rate was kept nearly constant at  $4 \times 10^9$  particles per spill (see table 1). The recovery of the light yield was examined during the day time between the two nights of irradiation.

### 2.1 Setup for the radiation hardness test

The experimental setup along the beam axis is shown in figure 1. The  $^{12}\text{C}$  beams were collimated within a 10 mm diameter spot by a 200 mm thick aluminum collimator placed at the downstream of the beam exit window. The number of beam particles passing through the GSO scintillator were counted by integrating the current from an ionization chamber (IC) that has two 2 mm air-gaps and was placed behind the collimator. The ionization chamber was also used to monitor the beam intensity not only to calculate the exposed dose of a GSO scintillator.

The beam profile was measured with monitor placed behind the ionization chamber. A 3 mm thick plastic scintillator was set on the beam axis for counting the number of particles in the low intensity beams for which the IC was not sensitive. A black box for GSO measurements was positioned downstream of the plastic scintillator.

Figure 2 shows the experimental setup inside the black box. There are two holes on along the beam axis on the front and back walls of the black box. The holes are sealed with a black plastic tape to avoid light leakage into the box. The energy of the  $^{12}\text{C}$  beam incident on the GSO scintillator was degraded from 290 MeV/n to 280 MeV/n by passing the plastic tapes, the scintillator, the



**Figure 2.** The experimental setup of GSO radiation hardness test inside the black box: a dashed line indicates the beam axis.

air and the other materials illustrated in figure 1 and figure 2. The energy deposit of  $^{12}\text{C}$  beam in the 1mm thick GSO scintillator is 52 MeV/ion (calculated based on Bethe-Bloch formula [7, 8]) or 56 MeV/ion (calculated by using GEANT4 [9]). In this study, 52MeV/ion was used to calculate exposed dose, and the 8% difference between two calculations is considered to be a systematic uncertainty of dose. Because the deposited energy is only 1.5% of the total energy, it is reasonable to assume the dose was uniform over 1 mm thickness along the beam. Two GSO scintillator plates ( $32\text{mm} \times 32\text{mm} \times 1\text{mm}^3$ ) were set on a movable stage (Sigma-Koki SGSP26-200: movable 0-200mm). One labeled GSO-R was placed on the beam axis and the other labeled GSO-L was placed 180mm away from the beam axis as shown in figure 2. The radiation hardness of GSO scintillator was evaluated by measuring its response to the very low intensity ( $10^3$  particles/spill)  $^{12}\text{C}$  beams (probe beam) for each GSO sample. The interval between of two successive spills was 3.3 sec. Particles were extracted for 1.2 sec during a spill. GSO light output was measured by two PMTs (Hamamatsu H1161 for the left "PMT-L" and Hamamatsu H6410 for the right "PMT-R") for redundancy. The response of GSO-R to the probe beam was measured immediately after high intensity irradiation was stopped. The GSO-L was moved to the beam axis by using the movable stage and the response of GSO-L to the probe beam was measured as a reference for no irradiation.

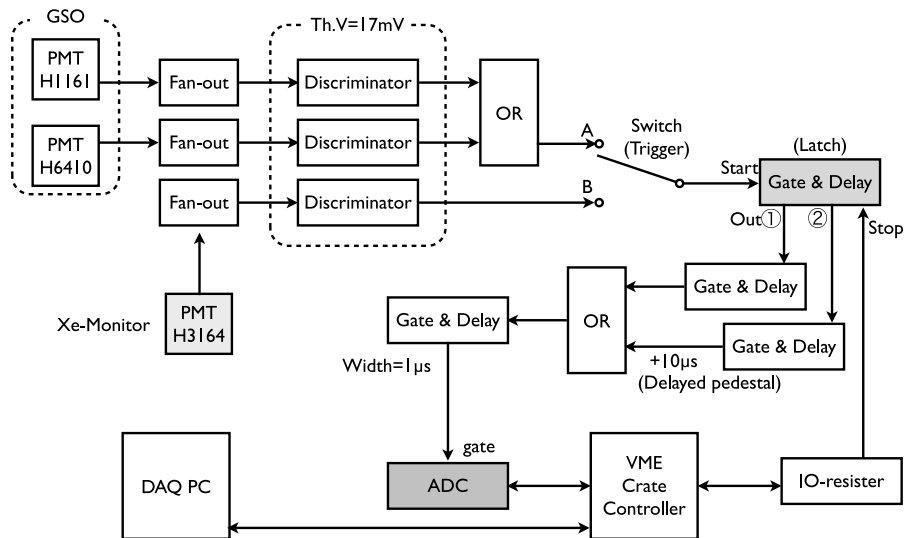
The measurements were carried out ten times as listed in table 1. The second and third columns show the intensity of the beam and the integrated dose to the GSO-R scintillator, respectively, calculated from the IC data. The fourth column shows the exposure time and the fifth column shows the dose rate. The runs from #0 to #5 were carried out in the first night and the runs from #6 to #9 were carried out in the second night.

## 2.2 DAQ setup

A diagram of the event trigger system (DAQ) is shown in figure 3. Two different types of trigger were used in the experiment. The GSO self trigger mode denoted as "Trigger A" was used for the measurements of the GSO response to the probe beam. The Xe flash lamp (explained in

Run#	Beam intensity [pps]	integrated dose [kGy]	exposure time[h]	dose rate [kGy/h]
0	-	0	-	-
1	$1.04 \times 10^7$	0.17	0.95	0.18
2	$1.18 \times 10^8$	1.77	0.80	2.01
3	$1.14 \times 10^9$	18.33	0.85	19.5
4	$2.80 \times 10^9$	60.76	0.88	48.2
5	$4.25 \times 10^9$	103.5	0.59	72.4
6	-	103.5	-	-
7	$4.40 \times 10^9$	243.5	1.86	75.3
8	$4.32 \times 10^9$	401.4	2.14	74.0
9	$4.46 \times 10^9$	684.4	3.70	76.4

**Table 1.** Radiation dose run information: Measurements were repeated 10 times. Each run corresponds to the plot point on figure. The unit [pps] means particles per spill. The runs #0 and #6 were carried out before starting irradiation and at the beginning of the 2nd night, respectively.



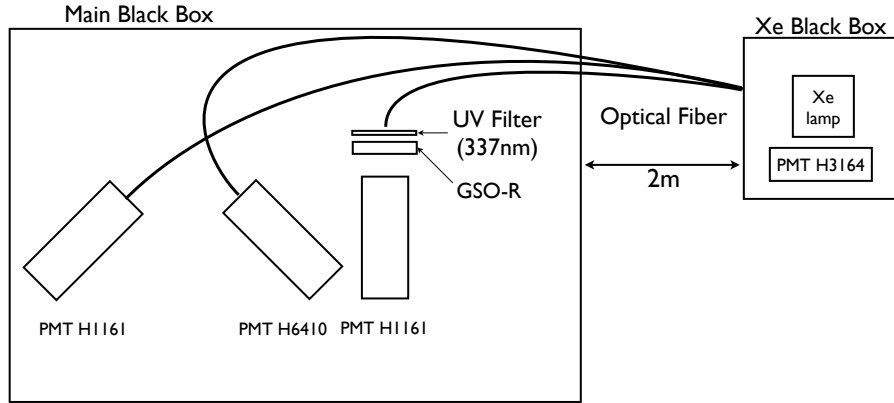
**Figure 3.** Block diagram of DAQ trigger system.

section 2.3) trigger denoted as “Trigger B” was used for those to the Xe flash lamp. These triggers were manually switched. The PMT pulses were measured by a CAEN V965A ADC module (800pC/12bit readout).

As shown in figure 3 the trigger signal was divided into two paths. One of them was used as a gate signal for the signal and the second as a gate signal for the event-by-event measurement of the pedestal after a 10  $\mu$ sec delay.

### 2.3 Setup for the Xe flash lamp test

It was observed that once the irradiation to the GSO scintillator was stopped, the light yield would recover to its un-irradiated value with the passage of time. The recovery was measured during the



**Figure 4.** Setup of the recovery test; The light from Xe flash lamp was fed to PMT and GSO-R with optical fiber.

day between the two nights of irradiation. A Xe flash lamp (Hamamatsu L4633C) was used for this measurement. The lamp was set in the second black box next to the experimental black box with a monitor PMT (Hamamatsu H3164).

Figure 4 shows the experimental setup for measurement of light yield recovery. The two black boxes were connected with three optical quartz fibers. The GSO samples were translated with the movable stage to position the GSO-R sample under a UV transmitting filter ( $337^{+6}_{-4}$  nm, FWHM) at the end of one of the optical fibers from the Xe flash lamp. The remaining two optical fibers were connected to the PMTs used in the irradiation test and are here used as intensity monitors of the Xe flash lamp. It had previously been confirmed that 337 nm UV-light can directly excite the GSO scintillator. Its response was measured by PMT Hamamatsu H1161.

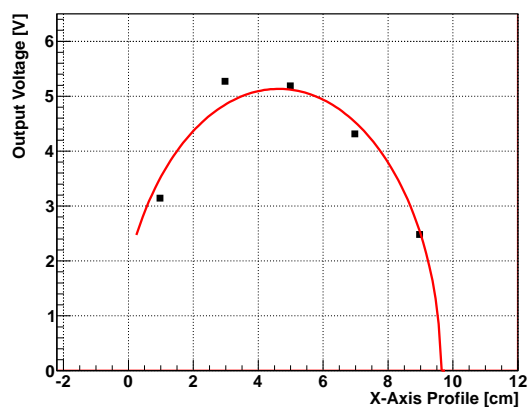
### 3 Results

#### 3.1 Data

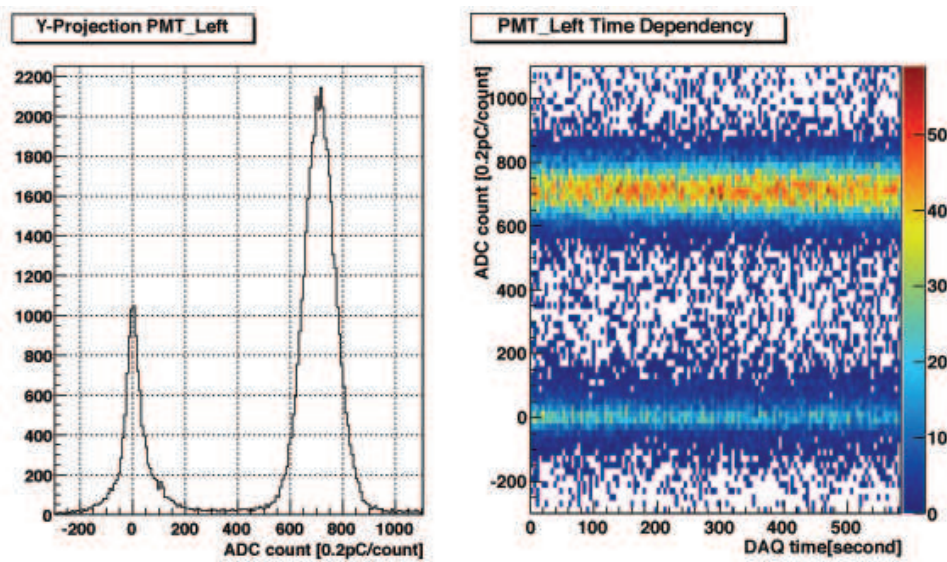
The radiation dose was calculated from the number of Carbon ions entering the GSO scintillator. This number is obtained by integrating the IC current. Black dots in figure 5 show a sample of beam profile obtained by the profile monitor. Because the beam profile is a projection of beam intensity in one direction, the expected profile for a uniform beam truncated by a collimator of radius  $r$  centered at  $x_0$  can be expressed as

$$f(x) = A\sqrt{r^2 - (x - x_0)^2} \quad (3.1)$$

where  $A$  is a normalization parameter and  $r = 5$  mm is the collimator radius. A best fit of equation (3.1) to the data is shown in figure 5 as a red curve. A reasonable agreement between the data and equation (3.1) is obtained. To simplify the analysis and discussion, the exposed dose is calculated in this study assuming a uniform beam profile.



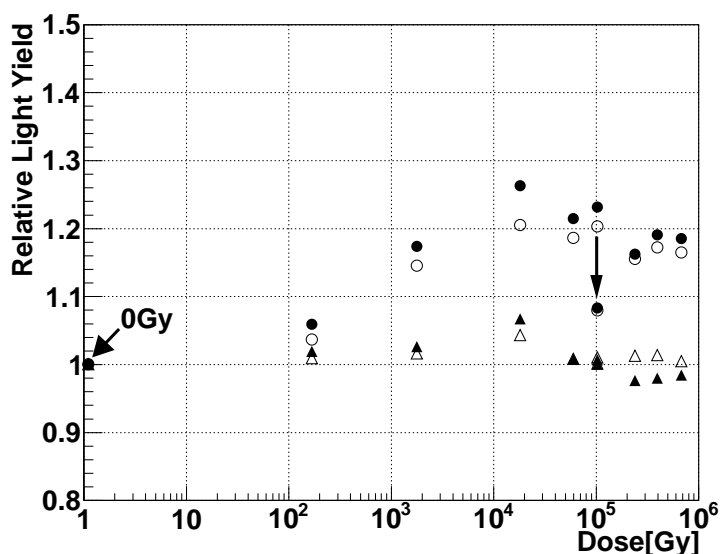
**Figure 5.** A sample of beam profile data. Black dots show data from the beam profile monitor. The red curve shows the result of fitting the data with a function that assumes a uniform beam intensity truncated by a collimator. A reasonable agreement between the data and the fitted function is obtained.



**Figure 6.** A typical result of the GSO response to the for probe beam. The left histogram is a time-integrated result of the right 2D histogram. The right peak in the left histogram is the Carbon peak (Signal) and the left is accidental trigger peak (Noise). The right panel shows the time dependence of the ADC data in a 2D histogram Color scale indicates the number of event in each bin.

Figure 6 shows a typical GSO response to the probe beam. The right panel shows the time dependence of the ADC data in a 2D histogram, and the left panel is the time-integrated ADC histogram. Color scale in the right histogram indicates the number of event in each bin. As discussed in section 3.3, the peak value of the GSO response changes with the time during exposure to the low intensity probe beam. Because of this change (even if it's only a few % shift), only data within 3 minutes from the start of exposure to the probe beam were used to determine the precise peak value of GSO light yield. In this criteria, statistical uncertainty was less than 0.1 % for each data.





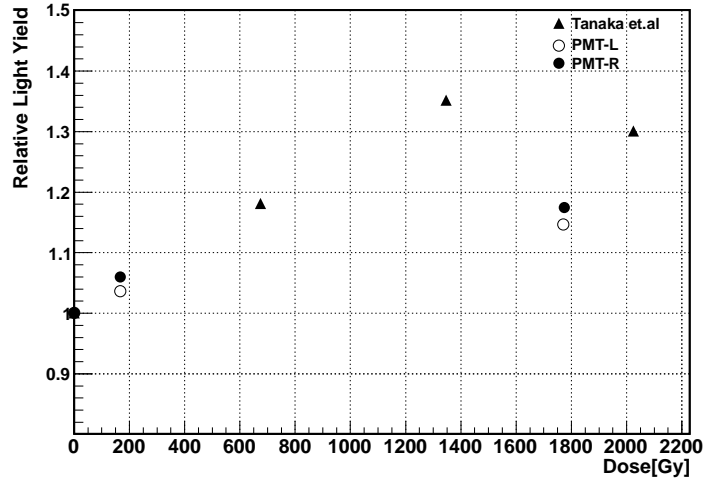
**Figure 7.** The relative light yield of GSO as a function of absorbed dose to GSO-R. The results are normalized to the first measurement (run #0) where the total dose is 0 Gy, while they are plotted at 1 Gy for the convenience on the logarithmic scale. The closed (open) markers indicate the output of PMT-L (PMT-R). The circle (triangle) markers indicate the light yield of GSO-R (GSO-L). The arrow at  $10^5$  Gy shows a decrease in light yield (recovery) during the day time.

### 3.2 Result of radiation hardness

GSO-R has received a total dose of  $7 \times 10^5$  Gy in this experiment. The relative light yield as a function of dose is shown in figure 7. The closed markers indicate the output of PMT-L while the open markers indicate that of PMT-R. Systematic uncertainty in this measurement is defined as  $\pm 3\%$  from the maximum difference between the outputs of PMT-L and PMT-R. This is far larger than the statistical uncertainty in determining the relative light yield. The circle (triangle) markers indicate the light yield of GSO-R (GSO-L). An arrow near  $10^5$  Gy indicates the 10% of recovery during the day between the two experimental nights of experiments (see section 3.3 for details).

No decrease but rather an increase in light yield was observed with increased exposure. The amount of increase reached a maximum of 25%. The increase seems to be related to the total dose below  $2 \times 10^4$  Gy, but not above. Only the output of the irradiated sample, GSO-R, shows increasing yield while the output of the reference sample, GSO-L, did not. Even considering the systematic uncertainty described above the output of GSO-R is significantly increased by exposure to irradiation.

A similar increase was also reported in a previous measurement using a  $^{60}\text{Co}$  gamma-ray source [2]. According to the result of [2], the increase was proportional to the irradiation dose at least below 1.5 kGy. Figure 8 is a close-up of figure 7 GSO data at low dose below 1800 Gy together with the results from the previous  $^{60}\text{Co}$  experiment [2] indicated by triangles. In the lower dose below 1.4 kGy, a good agreement with the previous gamma-ray result with a coefficient of proportion of 0.025%/Gy is found. At higher doses, however, the increase in the light yield seems to be saturated in both measurements.



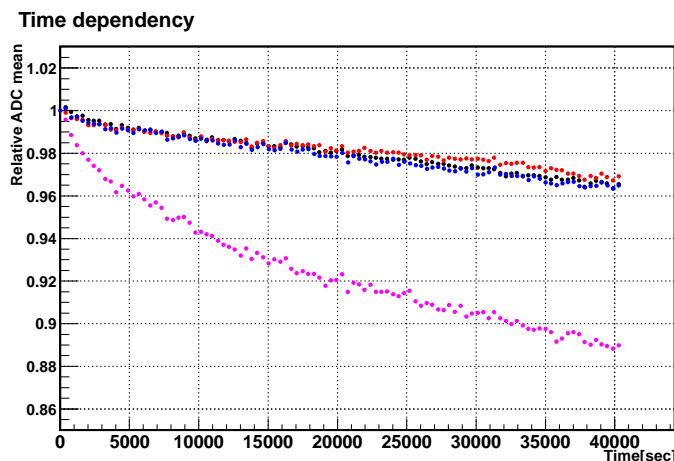
**Figure 8.** A close-up of figure 7 at low radiation dose but only for GSO-R. Triangle markers show the result from the gamma-ray exposure experiment [2].

In a future  $\sqrt{s} = 14$  TeV run of LHC, during the minimum running time required to accumulate the needed statistics for LHCf (about  $10\text{nb}^{-1}$ ), the detectors will receive of the order 100 Gy of irradiation. In this case the maximum increase in light yield of GSO scintillator is estimated to be about 2.5% without considering recovery during beam on and off times. Since the peak irradiation rate on the LHCf detectors is expected to be  $\sim 1.5$  Gy/h when the luminosity is about  $10^{29}$   $\text{cm}^{-2}\text{s}^{-1}$  in which period LHCf intends to take data, 60 hours of measurement is required. With this time scale the recovery may play a role and the increase in light yield will be suppressed compared to the Carbon ion experiments with the same radiation exposure even if a continuous measurement is performed. In case the luminosity is relatively high ( $L > 10^{31}$ ), however, the increase is not negligible. The increase is expected to be a few %/hour even if recovery is considered. Therefore, in this case it is necessary to calibrate the detectors by using some methods such as  $\pi^0$  mass reconstruction or  $\text{N}_2$  laser pulses.

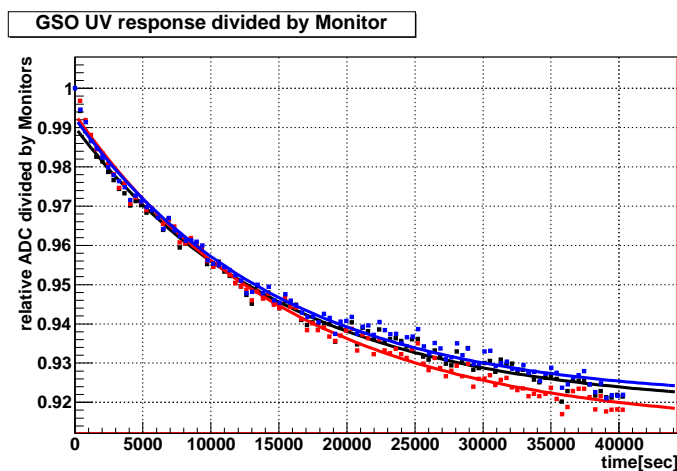
The cause of increase in the light yield of GSO has been discussed as follows [2, 10]. GSO scintillator has certain number of intermediate energy levels due to impurities or host ions in the energy gap that usually absorb the scintillation light emission. If the electrons generated by irradiation occupy these energy levels, then the absorb of scintillation light decrease and as a result, the light yield increases. The fact that our Carbon ion irradiation study obtained a result that is similar to gamma-ray irradiation indicates that an electronic effect is dominant in GSO over a possible nuclear reaction effect. It is expected that an increase in light yield due of occupation of the intermediate electron energy levels would have a decay life time when the irradiation is terminated as the electrons decay back to their ground state.

### 3.3 Result of recovery test

Figure 9 shows the time dependence of each PMT output to the Xe flash lamp irradiation during the day time. The horizontal axis indicates the elapsed time from the start in second and the vertical

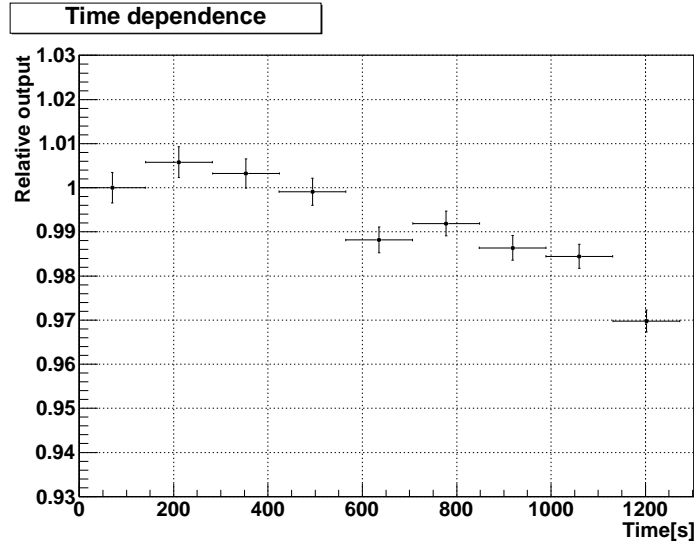


**Figure 9.** Variation of intensity of the Xe lamp and the GSO-R response to Xe's UV. Black, Red and Blue show the outputs of the monitors. Magenta shows the output of GSO.



**Figure 10.** Relative light output from GSO-R during the recovery test. Plots and curves with three different colors indicate the results corrected for the Xe lamp variation by using three different PMTs. Black (Red) corresponds to the signal of the GSO divided by PMT-L (PMT-R) and blue corresponds to that divided by the Xe-monitor PMT.

axis indicates the output of the PMTs relative to the output at the start time. Black, red and blue markers show the monitor outputs and they indicate the variation of the intensity of the Xe lamp. Black (red) corresponds to the output of PMT-L (PMT-R) and blue corresponds to the output of the Xe-monitor PMT. Magenta shows the light output of GSO excited by Xe lamp's UV component. While the intensity of the lamp itself has gradually decreased by 3.5%, the decrease in the GSO output by 11% is clearly larger and it indicates the evidence of recovery. The amplitude of recovery is consistent with the result obtained in the probe beam test as shown by the arrow in figure 7. To correct for the intensity variation of the Xe lamp, the light output of GSO-R was divided by each of the monitor outputs as shown in figure 10.



**Figure 11.** Short time scale recovery; Vertical axis is relative light yield.

To estimate the recovery time scale ( $\tau$ ), the data points in figure 10 were fitted with a function,

$$f(t) = A \exp\left(-\frac{t}{\tau}\right) + C. \quad (3.2)$$

The results are shown in figure 10 as three colored curves (black, red, blue) according to the monitor PMT used in the correction. The time constant  $\tau$  (presumably the life time of electrons in intermediate energy state) determined by the fitting is

$$\tau = 1.46 \times 10^4 \sim 1.59 \times 10^4 \text{ seconds} \quad (3.3)$$

together with  $A = 0.072 \sim 0.080$  and  $C = 0.914 \sim 0.921$ . The decay time scale  $\tau$  corresponds to about 4 hours and is long enough compared with our aforementioned probe beam measurements in three minutes that no correction to the probe beam measurements due to recovery is needed.

A possible faster recovery time scale was also observed during the run measuring the response to the probe beams just after radiation exposure. Figure 11 shows the time dependency of light yield of GSO measured by PMT-L after the irradiation Run #9. Mean values were obtained in nine time intervals. From this result, a few % of decrease in light yield was observed in 20 minutes, much faster than the time scale of equation (3.3). The deviation from the fitted equation (3.2) observed in figure 10 at times shorter than  $2 \times 10^3$  sec in figure 10 is also evidence for the existence of a fast component.

#### 4 Summary and Discussion

The radiation hardness of GSO (Ce:0.4%/mol) scintillator was tested by using Carbon beam in HIMAC. After exposure of  $7 \times 10^5$  Gy, the light yield of GSO scintillator did not decrease, but rather an increase up to about 25% was observed. The results are summarized in table 2 together with other measurements.

Measurement	Source	Dose [Gy]	Transmittance	Light yield	Ce [%/mol]
This study	$^{12}\text{C}$ (3.48GeV)	$10^6$	–	increase	0.4
A [2]	$\gamma$ from $^{60}\text{Co}$	$10^3$	decrease	increase moderate increase	0.5 1.5
B [3]	$\gamma$ from $^{60}\text{Co}$	$10^7$	no	–	0.5, 2.5
C [4]	$\gamma$ from $^{60}\text{Co}$	$10^5$	decrease	–	–
D [5]	p (12MeV)	$10^5$	decrease	–	0.5
E [6]	$^{12}\text{C}$ (50MeV)	$10^7$	decrease	decrease	1.5

**Table 2.** Summary of various measurements: The fourth column shows the change of transmittance of GSO scintillator. The fifth column shows the change of light yield. The sixth column shows the concentrations of Ce impurities in GSO.

The increase of light yield found in this study using a high energy Carbon beam is consistent with the previous study A carried out using  $^{60}\text{Co}$  gamma-ray source [2]. In the measurement E, however, no increase but a slight decrease in light yield up to  $7 \times 10^5$  Gy and a large decrease afterward were observed [6]. An anti correlation between the amplitude of increase in light yield and concentration of Ce impurities was also reported in A. Because the concentration of Ce impurities in GSO used in the measurement E and this study were 1.5%/mol and 0.4%/mol, respectively, this could partly explain the different consequences. Another notable difference between this study and E is the beam energy. Though in this study the beam lost 50 MeV uniformly along the 1 mm thick GSO, in the study E 50 MeV was lost only within  $35 \mu\text{m}$ . This 30 times different ionization density may cause a different response of the scintillators. The increase in light yield is initially proportional to the total dose but seems to be saturated above a radiation exposure of 1 kGy. The saturation was also previously observed in A. The recovery of the light yield was also observed in this study and the recovery time scale was estimated to be about  $1.5 \times 10^4$  seconds with another faster component.

## Acknowledgments

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