Afterglow Measurements of Lead Tungstate Crystals

H.Hofer, P.Lecomte, F.Nessi-Tedaldi

Swiss Federal Institute of Technology (ETH), CH-8093 Zürich, Switzerland

ABSTRACT

Following the observation of an excess of single photoelectrons in the scintillation spectrum of some lead tungstate crystals after irradiation, a method has been developed and applied to crystals grown under various conditions to determine whether scintillating lead tungstate crystals also respond to γ irradiation with long term afterglow, in the minutes to hours range.

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1. INTRODUCTION

The Compact Muon Solenoid (CMS) collaboration at the CERN Large Hadron Collider has selected lead tungstate crystals for its electromagnetic calorimeter (ECAL). Such a calorimeter must withstand a hostile radiation environment with radiation levels as high as 0.3Gy/h at the electromagnetic shower maximum in the ECAL barrel, and up to 15 Gy/h for $\eta=3$ in the end caps at the highest design luminosity of $10^{34} \text{ cm}^{-2}\text{s}^{-1}$ [1]. It has been established elsewhere [2] that electromagnetic radiation does not change the emission spectrum nor the scintillation decay kinetics but that it reduces the crystals transmission through the formation of color centers. However other effects can occur, such as long term emission of photons; their influence on the detector performance has to be understood and their acceptable levels for detector operation must be defined. In this paper we present a method to detect delayed photon emission which we illustrate with measurements on several crystals of different behaviors. We reported in a previous paper [3] on radiation hardness tests of crystals in preparation for the ECAL construction; the same irradiation procedure was used for the present studies.

2. TIME STRUCTURE OF LIGHT EMISSION IN LEAD TUNGSTATE CRYSTALS

Scintillation at room temperature is fast in lead tungstate [4],[5], but may in some cases include components with a few hundred nanoseconds duration, attributed at least in part to molybdenum contamination [2],[6] and eliminated by improving raw materials. Slow components can be observed for example with the so-called start-stop method [7]; however the precision and sensitivity of the method for long time constants are limited. Another way to observe slow components in the scintillation light is to digitize individual scintillation pulses; when the light level is low, each photoelectron can be distinguished as a few nanoseconds wide pulse, this width being defined mainly by the photomultiplier performance; for higher light intensity, these pulses merge and the envelope of the resulting signal shows the various scintillation components. Such a method works best when the number of photoelectrons produced from the single light pulse is large, so that the pulse appears as a smooth envelope of all emitted photoelectrons. Thus with lead tungstate, the method is preferably used in high energy particle beams of several GeV energy and served to detect slow components later attributed to Mo contamination as well as to measure the time response uniformity of some crystals [8]. The method is illustrated in **fig.1** where a single light pulse detected with a XP2262B photomulti-

plier [9] and generated by a 50GeV electron hitting a 23cm long lead tungstate crystal of hexagonal cross section, 1.5cm on a side, is shown (inverted for data analysis). The signal was fed to a digital oscilloscope LeCroy 9360 [10] via a high quality cable. The digitized data were then transferred to a UNIX system for analysis. Offline, the fitting of exponentials was performed, starting from the end of the pulse, through successive subtractions of the contributions from longer time constants, using the PAW analysis package [11].

Phosphorescence, that is continued light emission after the end of UV excitation, has been reported [12] with time constants in the range 15ms-127ms and conjectured to be also due to molybdenum contamination. Excitation of crystals with a pulsed UV light source was used to study the time constants in the range of up to a few ms [13].

Afterglow refers to the long term emission of light after irradiation, usually with γ , and we distinguish it from the above-mentioned slow components in the scintillation light because, owing to their long delays, the afterglow photons cannot be associated anymore with a specific incoming particle. It is also distinguished from phosphorescence since the excitation method is different. Afterglow with a time constant of 12min has been seen earlier in small undoped lead tungstate samples irradiated at massive doserates (between 40 and 1000 Gy/h) by counting the number of photoelectrons in a random gate of 100 ns duration [14]. Afterglow has also been reported in [2] using the anode current measurement method, with two time constants of ~6min and 42min. A similar method, monitoring the anode current from the silicon diode of a Hybrid PhotoMultiplier Tube (HPMT) is described in [15]. The first method lacks sensitivity whereas the second and third methods may give misleading results because the measured current can be due to a superposition of light **increase** due to afterglow and light **decrease** due to radiation damage. The anode current changes versus time do not yield sufficient information to separate the two opposite signs contributions. Transmission measurements during irradiation usually suffer from a similar limitation because the photomultiplier does not distinguish the scintillation light from the light of the monitoring source. The start-stop method commonly used to observe slow components in the scintillation emission is not suited to study afterglow because of the loss of correlation with the excitation, although afterglow may appear as a shift in the level of random stops, as reported in [13].

3. THE ENERGY SPECTRUM WITH ^{60}Co

We irradiate crystals using a 40 TBq ^{60}Co source. A weak ^{60}Co source of approximately 3 MBq placed on the tip of the crystal is used to excite its scintillation for light yield measurements. The crystal is mounted on a Philips XP2262B bialkali photocathode photomultiplier equipped with a standard CERN base type 4238. During irradiation, the photomultiplier high voltage is turned off, but it is turned on a few seconds after the irradiation is stopped, so that the crystal response can be monitored immediately and its changes tracked. The system reaches a gain stability of better than 1% within a few minutes after turn-on. The anode signal is split, and half of it goes to a charge integrating LeCroy 2249 ADC, the other half being used for triggering, through a threshold discriminator and gate generator usually set at 200ns width. Details of the setup can be found in [3]. The discriminator threshold is set at a level that allows the observation of the valley between the quasi-exponential decrease of the multiplier noise and the single photoelectron peak. An example of the spectrum observed can be seen in **fig.2a**, where the two-photoelectrons peak can also be distinguished, due to the good photomultiplier resolution and to the low ratio of the single-photoelectron rate to the multiple-photoelectrons scintillation rate from the γ hitting the crystal. The single photoelectron peak-to-valley ratio

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and peak position allow to monitor the photomultiplier stability [16]. Furthermore, capturing the entire single photoelectron peak enables the measurement of afterglow, which manifests itself as single photons emitted at random times by the crystal.

After irradiation, changes in crystals light yield are monitored by observing a displacement in the energy spectrum of the scintillation light produced in the crystal by the weak ^{60}Co source. An example is given in **fig.2**. During these measurements, we noticed that some crystals showed an excess of single photoelectrons in the spectra taken just after irradiation; **fig.3** show an example of such a behavior, with spectra taken respectively before, 3min after and 70min after irradiation. The observed excess is not an artifact of our test setup since other crystals (**fig. 2**) do not exhibit the same effect. This excess complicates the fitting procedure used to evaluate the light yield of the crystal, particularly when this light yield is low.

4. EVIDENCE FOR AFTERGLOW

The crystals considered in the present afterglow study are listed in **Table 1**. They are truncated pyramids and all have a length of 23 cm with some variations in transverse cross-section. All crystals were grown by the Bridgman method, except #1966, which was grown by the Czokralski method. All irradiations were performed at a doserate of 0.4Gy/h.

TABLE 1. Parameters of the crystals tested for afterglow and tests results

Crystal #	Front size [mm]	Back size [mm]	length [mm]	Doping	Dose [Gy]	Rate before [kHz]	Rate after [kHz]	τ [min]
1966	20.4	23.7	231	La	0.4	43.5	47	3
1991	20.0	23.2	230	none	0.4	120	350	20
1995	20.5	23.9	230	none	0.2	48	350	20
1996	20.4	24.0	230	La	0.2	46	92	5
1998	22.9	26.9	233	none	0.2	44	340	25
1999	22.7	26.7	229	none	0.2	40	175	5

To better understand the excess of single photoelectrons, we did set up a scaler counting our triggers for one second every fifteen seconds with a dead time of one microsecond. The dead time ensures that single incoming γ from ^{60}Co are not double counted in the scaler if the scintillation emission of the crystal is reasonably fast. When a γ excites scintillation in the crystal, one expects single photons to be emitted over time according to a distribution given by the shape of the scintillation pulse. If the crystal emits light uncorrelated to incoming γ , it will appear as an excess counting rate at the single photoelectron level. The rate before irradiation, which can be estimated from the monitoring source intensity and from the setup geometry, is around 40 to 50kHz, whereas the observed rate varies from crystal to crystal between 40 and 120 kHz. The contribution from photomultiplier noise has been checked separately and amounts to typically 2-3kHz 5min after power up.

Fig.4 shows the rate behavior after irradiation at 0.4 Gy/h for a La-doped, Czokralski-grown crystal (#1966); the measurement was performed at the end of a standard irradiation cycle in which the crystal received a total of 3Gy, the last 0.4Gy irradiation being uninterrupted. The rate is almost the same as before irradiation. The slight excess initially observed is compatible

FIGURE 8. Counting rate with calibration source after the end of irradiation for crystal 1996.

FIGURE 9. Counting rate with calibration source after the end of irradiation for crystal 1998.

FIGURE 10. Counting rate with calibration source after the end of irradiation for crystal 1999.

with the settling of the photomultiplier noise after high voltage turn-on. This shows that no sizeable afterglow is observed in this crystal, which we use as a reference, and that the photomultiplier rate settling is almost negligible.

The evolution of the rate as a function of time after the end of irradiation for the other crystals can be found in **fig.5 to fig.10**; data are uncorrected for dead time effects. The drop over time of the afterglow rate is generally not exponential, but we define for convenience in **table 1** a characteristic time as the one it takes for the excess rate to drop by a factor $1/e$. One observes that three out of four undoped Bridgman-grown crystals show a similar behavior, both in rate magnitude and in recovery time (of 20 to 25 min). Unlike them, crystal #1996, which is Lanthanum-doped, and crystal #1999, although undoped, show a less dramatic afterglow rate, which moreover drops faster, over 5 min. In all cases, the rates decrease non-exponentially to their value before irradiation within 1 to 2h and the spectra recover accordingly. Crystal #1991 also exhibits in addition an excess rate before irradiation. Since these measurements have been performed for full size crystals, the afterglow rate can be converted into a noise level for calorimeter operation. Using a typical detected crystal light yield of 10 photoelectrons/MeV, in a 200 ns gate one expects right at the end of irradiation a noise lower than 1 keV per crystal, which is well below the tolerable level in the final detector.

5. POSSIBLE CAUSES OF AFTERGLOW

Three years ago, we observed in a high energy electron beam that some crystals had slow scintillation components in the few hundred nanoseconds range [8], which were later attributed to impurities in the raw materials, chiefly molybdenum [2],[6]. Although the time domain involved here is completely different, we suspected a similar cause and checked the corresponding crystals for impurities. Glow Discharge Mass Spectrometry (GDMS) analysis of two samples per crystal, one at each extremity, was performed for us by Shiva technologies [17]. Aside from #1996, all the crystals were satisfactory in terms of impurities (**Table 2**) except possibly for some traces of lithium, sodium and potassium, although the lanthanum doped crystal showed strong dopant segregation.

TABLE 2. Major trace elements from GDMS analysis; the two values per element for each crystal correspond to the back and front extremities of the crystal (relevant numbers are in bold).

Crystal #	Li	Na	K	As	Nb	Mo	La
1995	1.1	8.4	7.9	1.2	0.19	1.5	1.9
	0.79	6.1	1.2	0.09	0.14	1.3	2.1
1996	2.7	10	1.1	1.1	<0.01	18	200
	1.6	3.1	0.90	2.8	<0.01	3.1	0.44
1998	2.3	3.1	1.3	2.3	<0.01	0.78	1.3
	4.7	0.28	0.30	0.41	0.02	0.66	2.7
1999	2.9	10	0.39	0.31	<0.01	1.3	1.0
	2.9	2.3	0.37	1.6	<0.01	0.37	0.49

The dopant could suppress afterglow and the small amount of afterglow left in #1996 could be attributed to the inhomogeneous dopant distribution in this Bridgman-grown crystal; unfortunately, we did not get samples from #1966 for GDMS analysis to test a possible relation between dopant segregation and amount of afterglow. The cause of the afterglow does not

FIGURE 5. Counting rates with calibration source after the end of irradiation for crystal 1991. Note that even before irradiation, the trigger rate is higher than for crystal 1966 (fig.4), although the source is used approximately in the same position. The time scale is unchanged between fig.4 and fig.5 but the vertical scale in fig.4 is magnified by a factor ten compared to the one of fig.5.

FIGURE 6. Counting rate with calibration source after the end of irradiation for crystal 1991 (same as on fig.5), until fully recovered.

FIGURE 7. Counting rate with calibration source after the end of irradiation for crystal 1995.

seem to be trace elements, so we are looking at other possible explanations, noting that we have up to now detected afterglow only in Bridgman-grown crystals (we have tested several other doped crystals grown by the Czokralski method and not listed here; none of them showed significant afterglow).

6. CONCLUSION

We have measured afterglow rates in crystals after irradiation through the detection of single photoelectrons from lead tungstate crystals. This technique is simple and can easily be applied to the study of other crystals. We observe radiation-induced afterglow rates which differ considerably from crystal to crystal. Their level is nevertheless well within the requirements for use in the CMS detector, at least in the time domain investigated. However, such an effect can be an indication of structural problems in the crystals, and we thus believe it is a parameter which has to be included in the quality control procedures for crystals acceptance.

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FIGURE 1. a) Digitized pulse from a single 50 GeV electron in a lead tungstate crystal.
b) Fit of the slow component (note the reflection around $t=270\text{ns}$, due to a missing back termination).
c) Fit of the fast component after subtraction of the slow component

FIGURE 2. Spectrum of crystal 1966: a) before irradiation. b) 3min after the end of irradiation.

FIGURE 3. Spectra of crystal 1998: a) before irradiation. b) 3min after the end of irradiation. The spectrum is completely dominated by single photoelectrons, a fact reflected in the increased trigger rate (see table 1). c) 70min after the end of irradiation. The single photoelectrons rate has decreased almost to the preirradiation level

FIGURE 4. Counting rates with calibration source after the end of irradiation for crystal 1966.