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Study of Radiation Damage in Lead Tungstate Crystals Using Intense High Energy Beams

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We report on the effects of radiation on the light output of lead tungstate crystals. The crystals were irradiated by pure, intense high energy electron and hadron beams as well as by a mixture of hadrons, neutrons and gammas. The crystals were manufactured in Bogoroditsk, Apatity (both Russia), and Shanghai (China). These studies were carried out at the 70-GeV proton accelerator in Protvino.

I. INTRODUCTION

The BTeV [1] experiment is being readied to study beauty and charm physics at the Fermilab Tevatron collider. The goals are to make an exhaustive search for physics beyond the Standard Model (SM) and make precise measurements of the SM parameters. The important measurements to make involve CP violation, mixing, and rare decays of hadrons containing b or c quarks. Since detection of photons, mostly from π^0 or η decays is essential to accomplish our physics objectives, we have decided to use an electromagnetic calorimeter (EMCAL) made of lead tungstate PbWO_4 (PWO) crystals. These crystals produce light proportional to the incident electromagnetic energy; this light will be sensed by photomultiplier tubes. This system is ideal for a heavy quark experiment at a hadron collider because of excellent energy and position resolution, a compact shower size that minimizes overlapping showers (due to the small Moliere radius), fast signals that minimize shower overlaps in time and expected excellent radiation hardness.

Pioneering work on PWO crystals performance was done at Protvino [2]. These results showed the promise of such crystals. However, the technology of mass producing such crystals with high purity was not yet known. The CMS group worked with companies both in Russia and China to perfect these techniques [3].

In high luminosity collider experiments, PWO crystals will be irradiated by high energy particles and accumulate significant absorbed doses, up to a few Mrad. The radiation hardness of PWO crystals has been studied by the CMS group using radioactive sources and electron beams [4]. The general conclusion is that lead tungstate crystals were radiation hard, and that the damage in crystals depends only on the dose rate [5]. It is, however, important to measure radiation damages of PWO crystals in high energy particle environments which are more similar to that which these crystals will be exposed to.

It is important to emphasize [6,7] that in a hadron collider experiment radiation effects from hadronic interactions and neutrons could be much more serious than seen with photons or electrons of the same doses.

*deceased

Compared to photons or electrons, high-energy hadrons will be able to induce inelastic nuclear reactions which will locally destroy the crystal lattice. In particular, they can create nuclear fragments with very high energy transfer and lead to extended clusters of crystal lattice distortion. A simple calculation suggests that such interaction may produce significant number of additional crystal defects over the life of BTeV. Therefore it is crucial to study the radiation hardness of PWO-crystals using a hadron environment which is similar to the BTeV EMCAL expectations. Such radiation studies with lead tungstate crystals have been carried out for the first time. The results of this study are presented in this paper.

The general goal of our test beam studies was to evaluate the performance of lead tungstate crystals produced by two manufacturers in Russia, Bogoroditsk and Northern Crystal in Apatity, and one in China, Shanghai Institute of Ceramics. More specific goals were to understand how to set specifications for purchasing crystals, confirm energy and position resolution predictions, measure the radiation rate dependence of light output, and measure the correlation between light output and the LED calibration system at varying radiation loads. The 2B beam channel at the Protvino accelerator U70 has been specifically developed to provide these measurements [8]. Results on energy and position resolutions of the PWO crystals which were obtained in these runs have been published elsewhere [9].

This paper is organized as follows. A general picture of radiation damage of PWO crystals as well as the results of simulations on dose rate profiles in the PWO crystals with the use of the MARS program [10] are described in Sec. II. These calculations are made for the BTeV experiment and for the two types of radiation studies of PWO crystals which have been carried out in Protvino for BTeV. In these studies we irradiated crystals with (a) moderate dose rates (1-60 rad/h) of high-intensity high-energy electron and pion beams in the secondary particle channel 2B and (b) super-intensive dose rates of mixed beam at a dedicated facility that was several meters away from the main ring of the U70. The test beam facility for approach (a), including phototube monitoring as well as the results of the moderate dose rates irradiation are discussed in Sec. III. Three accelerator runs, each up to a month long were devoted to these studies. The results from approach (b) are given in Sec. IV. The conclusions of the entire radiation studies are presented in Sec. V.

II. RADIATION DAMAGE AND ABSORBED DOSE PROFILES IN THE CRYSTALS

Radiation hardness studies of detectors and electronics are an important concern in EMCAL design [3,4] All crystal scintillators suffer from radiation damage. The most common radiation damage is due to color center formation, which results from trapping of electrons in crystal defects such as vacancies, displacements and impurities [11]. These electrons are often in metastable states and can be excited by visible photons to higher energies. Color centers reduce light transparency of crystals, resulting in reduced light output. Additional damage may be caused by hadrons when they create crystal defects by displacing nuclei or changing nuclei to different nuclei. This kind of damage can not only reduce light transparency, but, in principle, also reduce primary scintillation light itself. It would be more difficult to monitor the latter effect. Since the trapped electrons are in metastable states of varying lifetimes and “potential barriers”, some of them may disappear very quickly, whereas others may be almost permanent.

When a PWO crystal no longer receives radiation, its color centers (semi-stable excited states) disappear, and it recovers from transmission degradation by natural room-temperature annealing. In fact, this annealing goes on even during radiation exposure. In general the rate of radiation damage decreases with the amount of damage. Therefore, when crystals are exposed to a constant radiation level, they lose light only up to the point when the rates of radiation damage and natural recovery balance. Raised temperatures accelerate the recovery process and so may ultra violet irradiation. Because the damage may recover at room temperature, it leads to a dose rate dependence of the light output.

The CMS experimental data, mainly from photon and electron irradiation, indicate that the light transmission of crystals deteriorates due to formation of color centers by radiation, while the scintillation mechanism itself seems unaffected. Besides dependence on the dose rate, the radiation damage of PWO crystals could also be sensitive to the type of radiation. In particular, the properties of crystals could be significantly degraded in hadron beams by displacement damage effects, i.e. distortions of the crystal structure. In these studies it is very useful to know the hadron fluence, the hadron spectra and the absorbed dose rate.

The BTeV EMCAL extends radially outward from the beam line. The crystals near the beam pipe receive the maximum dose. In order to ascertain the level of radiation in the crystals we performed calculations using the MARS code. Results are given in Table I.

TABLE I. Fraction of BTeV crystals with given absorbed doses and dose rates estimated at the maximum of the dose profiles inside the crystals (100 rad = 1 Gy)

Fraction (%)	Absorbed dose (krad/year)	Dose rate (rad/h)
11	0.3 - 1	0.11 - 0.36
22	1 - 2	0.36 - 0.72
27	2 - 5	0.72 - 1.8
12	5 - 10	1.8 - 3.6
16	10 - 50	3.6 - 18
6	50 - 100	18 - 36
3	100 - 200	36 - 72
2	200 - 500	72 - 180
0.4	500 - 1000	180 - 360
0.2	1000 - 2000	360 - 720

We tried to emulate the BTeV conditions as much as possible. A 27 GeV electron beam and a 40 GeV π^- beam have been used to irradiate the crystals with moderate dose rates. The beams were directed into the secondary beam channel from the accelerator, where primary 70-GeV protons interacted with an internal target. The MARS calculations of the absorbed dose rates in the crystals from the secondary beam channel are compared with the absorbed dose rates expected in BTeV in Fig. 1. The η (pseudo-rapidity) shown here reflects the coverage of the BTeV EMCAL, where η of 4.45 is at the extreme inside near the beam and η of 2.27 is on the extreme outside. Electron and pion dose profiles in the crystals are different. The crystals receive damage from pions almost uniformly along their length starting from a distance of 5-7 cm from the front. For electrons an absorbed dose rate at shower maximum is two orders of magnitude higher than near the crystal ends. Because the BTeV dipole magnet sweeps particles vertically, the radiation profile at the calorimeter is different in the horizontal and vertical planes. Thus in BTeV the mix of charged hadrons and photons changes and the ratio between shower maximum and the crystal ends is only a few times in the vertical plane and an order of magnitude in the horizontal plane. That is why both electron and pion beams are used to study radiation damage of the crystals.

Two crystals, one manufactured in Bogoroditsk and the other in Shanghai were placed near the vacuum pipe of the Protvino U-70 accelerator in the first dedicated super-intensive dose rate study. These crystals were irradiated by secondary particles coming out the internal target of the accelerator. The energy spectra of neutrons, gamma-quanta and charged hadrons at the place where the crystals were irradiated are shown in Fig. 2(b). For comparison the expected particle spectra at the front face of the BTeV EMCAL are presented at the top part of the same Figure. We can see that the spectra look similar, although the dose rate in the IHEP irradiation zone is about three orders of magnitude higher than expected in BTeV. In the second dedicated intensive study, four more crystals from Bogoroditsk and Shanghai were exposed to radiation at the same facility. The intensity of the second run was reduced by two orders of magnitude. Absorbed dose rates as a function of longitudinal position for these two exposures are presented in Fig.3.

III. MODERATE DOSE RATE IRRADIATION

In this Section, we describe the testbeam facility for moderate dose rate irradiation studies, discuss phototube gain monitoring, and present the results of irradiating crystals with electrons and pions. In our

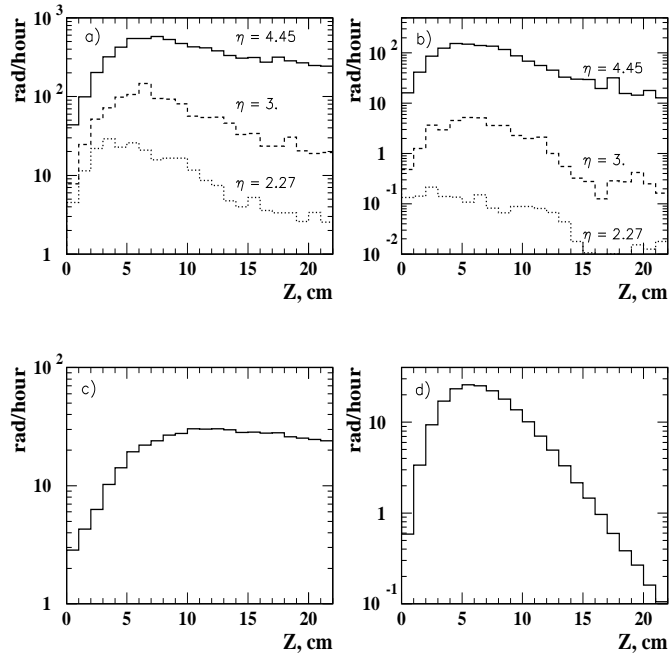


FIG. 1. Longitudinal profiles of the absorbed dose rate at the vertical (a) and horizontal (b) planes of the BTeV EMCAL at different rapidities, and at IHEP testbeam with 40 GeV pions (c) and 27 GeV electrons (d). The length of the crystal is 22 cm. The electron profile is normalized by $10^4 e^-/\text{sec}$, and the pion profile by $10^5 \pi/\text{sec}$.

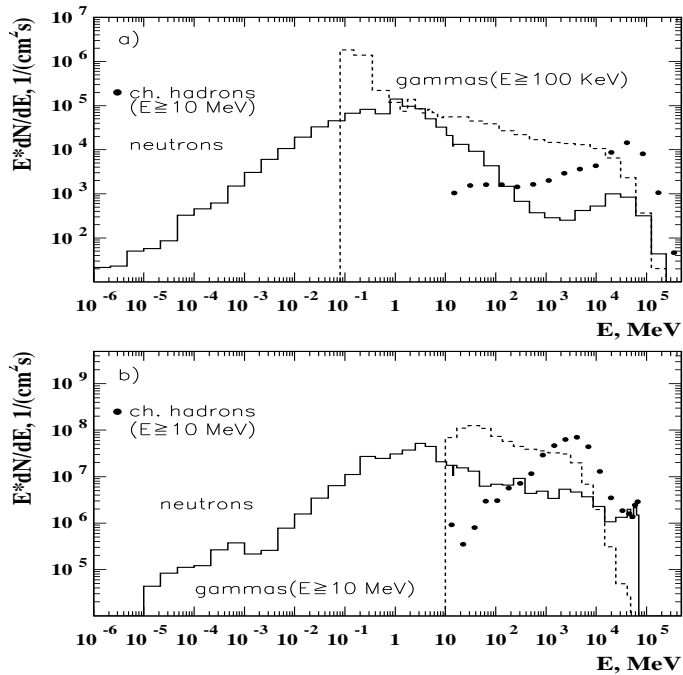


FIG. 2. (a) Particle spectra at the BTeV EMCAL. (b) Particle spectra in the dedicated superintensive dose zone near the vacuum ring of the U-70 accelerator. These spectrum shapes are very similar although (b) is about three orders of magnitude higher.

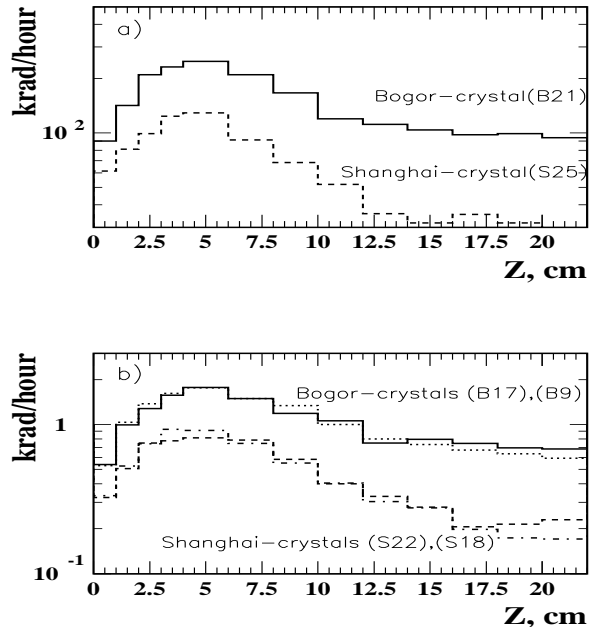


FIG. 3. Absorbed dose rates as a function of longitudinal position at the dedicated facility near the internal target 27 of the U-70 accelerator for two crystals in the exposure (a) and four other crystals in the exposure (b). The intensity of primary 70-GeV protons at the internal target in the second exposure was three orders of magnitude less than in the first one.

radiation studies we wanted to use radiation conditions as close to the BTeV conditions for the crystals as possible. Absorbed dose rates as a function of longitudinal profile at the BTeV EMCAL and at IHEP testbeam have been already discussed in Section II and presented in Fig. 1. We used 27-GeV electrons and 40-GeV pions to irradiate crystals in the three accelerator runs.

A. Test beam facility

The test beam setup consisted of 5x5 PWO crystal array situated inside a temperature controlled light-tight box (ECAL), a beam with a momentum tagging system and a scintillation counter trigger system [8], [9].

All the crystals we used were rectangular in shape. The Bogoroditsk and Shanghai crystals were 27×27 mm² in cross section and 220 mm in length. The Apatity crystals were 22×22 mm² in cross section and 180 mm in length. Light from each crystal was collected by a 10-stage 1-inch diameter Hamamatsu R5800 photomultiplier tube (PMT). All the crystals were wrapped by a 170 μ m thick tyvek. A radioactive source study at University of Minnesota showed that tyvek is radiation hard up to at least a few Mrad. This study as well as the Belarussian State University(Minsk) one also showed that a borosilicate glass did not lose any light at least up to 10 krad, a quartz glass up to 1 Mrad, both with an accuracy of 1%. Six quartz PMT's were used for a part of our test beam study, the rest were the borosilicate PMT's.

We accumulated absorbed doses in our crystals up to a few krad. No changes inside the box or PMT HV values were made during the irradiation period. The PWO light yield strongly depends on crystal temperature [3]. The 25 crystals were surrounded by a set of four copper plates that were water cooled, which enabled a temperature control using a Lauda cryothermostat. The temperature for the study described in this paper was fixed at $20^{\circ}C \pm 0.1^{\circ}C$. To measure the temperature of the crystals, 24 temperature sensors were mounted on the front and rear faces of the crystals.

For the most of the results presented in this paper, the crystal array was monitored with the four different

wave length light emitting diodes (LED). The LEDs had the following wavelengths : 660 nm (red), 580 nm (yellow), 530 nm (green), and 470 nm (blue). Transmission of red light in the crystals is not affected much by radiation damage [12], so the red LED monitors the PMT gain change. One LED generator with a multiplexer was placed into the light-tight box with the crystals and used for the all the LEDs. The LED temperature dependence is on average 1%/1°C, and thus limited to 0.1% because of our careful temperature control inside the box. We had one bunch of fibers between the LED generator and the crystals. In each accelerator cycle 10 pulses data for one LED color were collected. Four cycles were needed to collect all the LED signals.

An α -source (YAP-light pulser [13]) was mounted on the photocathode of a separate PMT in addition to the fiber to monitor LEDs themselves. It had 20 decays/sec with about 5,000 photons/pulse. Forty pulses were collected each spill. A signal from last dynode of this PMT was used to form an α -trigger. The size of the YAP crystal was $3 \times 3 \text{ mm}^2$ with thickness of 0.1 mm. Its emission spectrum has the maximum at 360 nm. The YAP crystal temperature dependence of the light output was 0.4%/1°C. The α -spectrum as well as α -stability is presented in Fig. 4. One can see that this stability over 85 hours is better than 0.2%. A Hamamatsu PIN diode S6468-05 with integrated amplifiers was also used to monitor the LEDs because it has a good sensitivity in the red region as well as a gain stability. It's temperature dependence is much less than 0.4%/1°C.

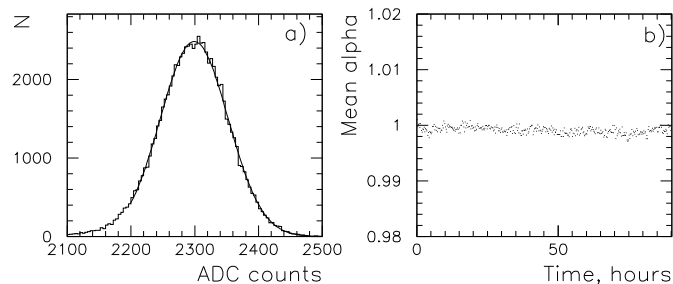


FIG. 4. (a) α -spectrum accumulated over 1.5 hours. Sigma/mean = 2.3% when it is fitted by a Gaussian. (b) Normalized α -signal in time to show α -stability over 85 hours. Each point corresponds to a 15-minute measurement.

We did not use an optical grease coupling between the crystals and the PMT's in order to avoid a contribution of a possible radiation damage of the grease. The PMT's were attached to the crystals without any optical material between them. High voltage to the tubes was supplied by a LeCroy 1440 HV system. Signals were sent to the control room patch-panel without any connection to ground inside the crystal box to avoid ground loops. A LeCroy 2285 15-bit integrating ADC was used to measure charge over 150 ns without pedestal subtraction. The ADC sensitivity was 30 fC per count. At HV values around 1000 V in the tubes we had about 2 MeV/ADC count.

B. Phototube gain change monitoring

We used high-intensity high-energy electron beam to irradiate the crystals and at the same time monitor the light output. The beam particles travel along the length of the crystals toward the PMT. We needed to take into account the possible phototubes gain changes, for example, from varying in the beam intensity. Thus, we carried out two types of PMT gain change studies to separate the effect of PMT gain change from crystal radiation damage. We investigated the possible changes in PMT gains at a dedicated stand at IHEP after the accelerator runs. We also monitored the PMT's continuously during one of the runs using the red LED.

Fig. 5 shows a schematics of the dedicated stand setup to study the PMT behavior, where the average anode current was adjustable by changing the intensity of DC light shining on the PMT. The setup consisted of a high quality referenced PMT(Hamamatsu R5900), a blue LED light pulser, a DC LED. Both pulsed and DC LED lights were injected into the test PMT through optical fibers. The stability of the pulsed LED itself was monitored by a *Pu* radioactive source implanted in a crystal and mounted at the photocathode of the reference PMT. The read-out and control electronics were placed in a CAMAC crate which had an interface with a PC. The average anode current was chosen for each test PMT to be the same as what we had at the test beam. The anode current was measured directly by an ammeter. Fig. 6 shows a timing diagram of various measurements. Each set of measurements took 2 minutes. At the beginning of each set, we measured the pulse heights of two groups of 2000 light pulses. It took 20 sec to collect 2000 pulse data and there was a 10-sec interval between the two groups. The data from a radioactive source in a self trigger mode were collected during the remaining 70 seconds. This 2-minute set was then continuously repeated. The intensity of the DC LED to induce a finite average anode current in the tested PMT was allowed to change, if needed, in the 10 second time intervals. This system allowed us to make PMT long-term stability measurements with a precision of 0.2%.

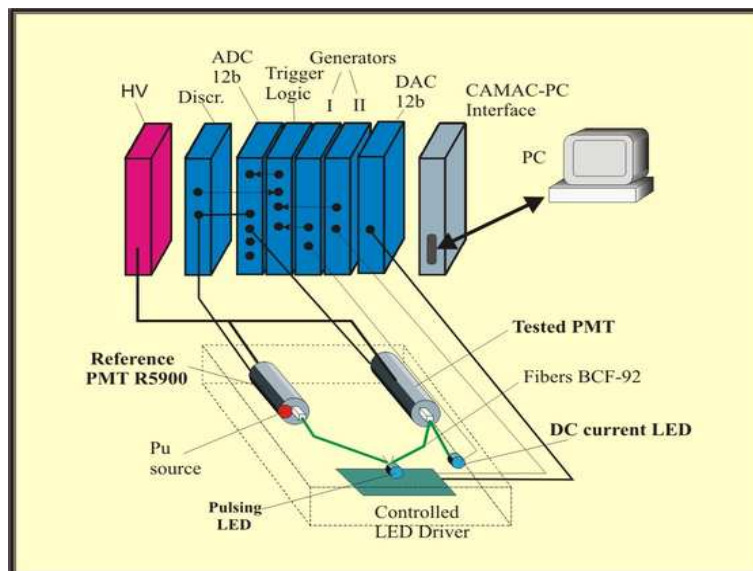


FIG. 5. Sketch of a dedicated stand setup to study a PMT gain variation.

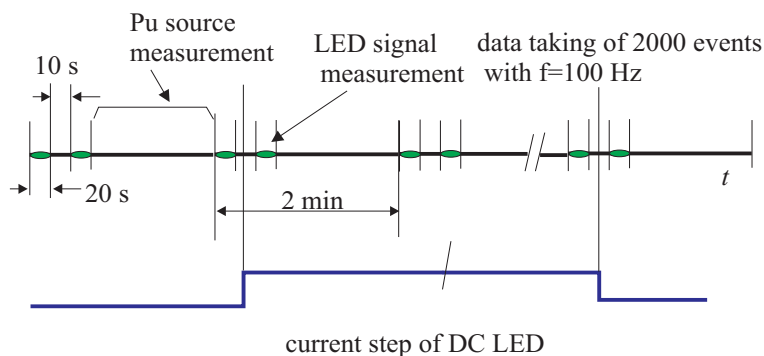


FIG. 6. A timing diagram of our test stand to measure PMT gain variations.

We used positive HV for PMT's with grounded photocathodes for the first accelerator run and negative HV for the second and the third runs. The red LED response of PMT number 743 using negative HV during irradiation study is presented in Fig. 7. The behavior of the same PMT at the dedicated stand is shown in

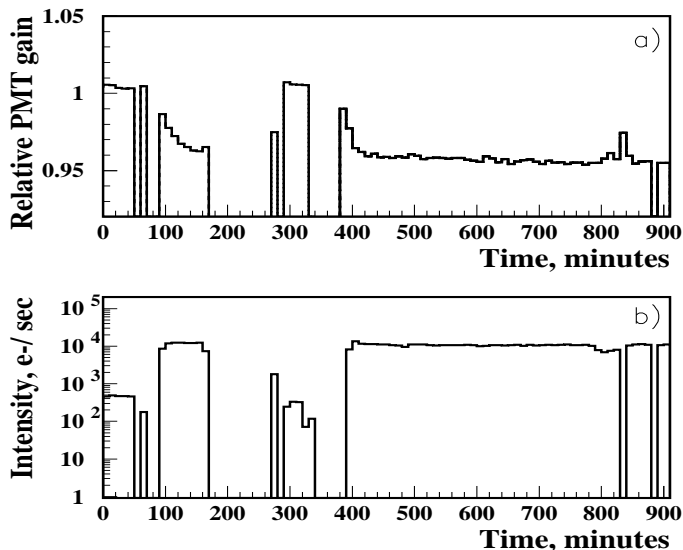


FIG. 7. (a) The response of PMT number 743 using a red LED for a central crystal in the array as a function of time. (b) Beam intensity in this counter as a function of time.

Fig. 8. We see that short-term loss of the signal is 3-5 % when the test beam intensity is at the level of 10^4 e^- /sec averaged over the entire accelerator cycle.

A similar signal loss was seen (Fig. 8) when the additional green LED was turned on to produce the anode current of $5 \mu A$. Another similarity is that when the “beam” or green LED was turned off, the PMT gain rose by a few percent. We compared the behavior of each PMT at the stand and during the beam test and found a satisfactory agreement between these results.

Six phototubes with the quartz glasses (Hamamatsu R5800Q) of the same size were used to avoid the possible radiation damage to phototube windows. These PMT’s had a gain change of 5-6% and one of them even 10%. The gain variation of one of these quartz phototubes is presented in Fig. 7. For the PMT’s with borosilicate glasses the signal loss has been measured not to exceed 3% for both positive and negative HV.

The blue LED signal amplitude over 85 hours is presented in Fig. 9. Fig. 9(a) shows the electron beam intensity over this time period in a sample crystal. Fig 9(b) shows the raw blue LED signal for the same crystal. We see that the blue LED signal fell by 5-6% when the beam was off. The time diagram of the blue LED corrected by the red LED is shown in Fig. 9(c). Note that most of our PMT’s lost gain when the beam was on. In our plot we selected this PMT with the opposite behaviour to show that we could correct for this big gain change even though the sign of the change was atypical.

All of our analyses included corrections using the red LED data. We corrected the signals from electrons and blue LED on the signal from red LED to subtract a PMT gain variation effect from the total signal for each PMT. When the green or yellow LED signals were used in the analysis, they also were corrected using the signal from red LED.

To check and correct the stability of the red LED, we used the α -source. The instability of blue, green and yellow LEDs was corrected using the PIN diode. The ratio of the PIN to α signals was stable to an accuracy of 0.1 %. To decrease any possible remaining LED instability left after these corrections, we kept for further analysis only accelerator spills with similar beam intensity. We conservatively estimate that the error for the blue LED signal is 0.2%.

C. Irradiation by high-energy electrons

The crystal array was irradiated by 27 GeV electrons for one week with an accelerator efficiency of 85%. The beam intensity at the crystal array was 6×10^5 particles/spill most of the time during this period. The 80% of the beam entered in one of the six central crystals. About a half of the time, the beam was centered on one crystal in the array and during the rest of the time it was centered on another crystal. Coordinates of the electrons entering the crystal array were measured by the drift chambers. The events with electrons near the center of the crystals were selected for data analysis.

We now describe the analysis of the electron beam data. All the information which was accumulated during 85 beam hours (one position of the beam at the array, see above) was divided into pieces of two hours long each. This choice was made to have enough statistics to measure the average energy deposit in a crystal with an accuracy of 0.3%, and thus we could continuously monitor the crystal signal loss. Prior to the irradiation study, the PMT gain of the each crystal in the array was adjusted to 10,000 ADC counts when 27 GeV electron hit the center of the crystal. Since this corresponds to 76% of the full electron energy [9], one ADC count corresponded to 2 MeV. The size of the beam spot was chosen $4 \times 4 \text{ mm}^2$ for most irradiated crystals and $6 \times 6 \text{ mm}^2$ for crystals with lower doses in order to equalize the statistics. The true coordinates of a particle at the array was calculated with the information from the last drift chamber which was close to the array. The accumulated energy peaks were fitted by a Gaussian. Then the mean values were corrected using the red LED.

For each crystal a dose rate was defined as an effective number N of electrons per second hitting this crystal multiplied by $25.9 \cdot 10^{-4}$ (see Fig. 1(d)). The number N was calculated as energy deposit in this crystal in GeV/sec divided by 20.5 GeV (it corresponds to 76% of 27 GeV energy deposit when electron hits the center of the crystal [9] in accordance with the MARS simulation).

A typical result for an irradiated crystal is presented in Fig. 10. In Fig. 10(b) we see an intensity of the electron beam which is shown in dose rate units at the shower maximum according to the MARS simulation results presented in Fig. 1. The absorbed dose is given in Fig. 10(c). The main result is shown in Fig. 10(a) which is the normalized electron signal. We see that finally the crystal lost 12% of the signal under an electron beam irradiation mostly with 15 rad/h dose rate after it accumulated 1.2 krad absorbed dose. It also appears that the radiation damage is saturating. For dose rates of 10-25 rad/h under 27 GeV electron beam irradiation, eight crystals lost an average of 8% after a total accumulated dose of 1-2 krad.

In order to use the light monitoring system to track the effects of radiation damage, it is necessary to determine the relation between the change observed by the monitoring system and the change in the signal from beam electrons. Because of the different optical paths taken by the injected monitoring light as compared to the scintillation light this constant is not expected to be unity. Furthermore the LED system monitors the transparency of the crystal at a specific wavelengths and thus does not sample the entire spectrum of scintillation light.

The blue LED emits at 470 nm and the scintillation peak is at 430 nm. The typical blue LED and electron signal behavior under irradiation for one of the crystals is shown in Fig. 11(a). The blue LED (as well as the electron signal) is corrected by the red LED, and the red LED by the α -source. The same was provided for the green and yellow LEDs. For the green LED a signal loss was smaller than for the blue LED, and for the yellow LED the signal loss was smaller still (not shown). In Fig. 11(b) we see a strong correlation between the change in the blue LED light level and the beam signal. We fit such distributions by the straight lines, ignoring some deviations from linearity. The results for a few crystals are presented in Fig. 11(c). We did not observe a significant difference in the crystals from different manufacturers. Constants of proportionality vary from 0.3 to 0.6 for these crystals. The dependence of a relative electron signal on the absorbed dose is presented in Fig. 12.

A simple model is used to describe signal loss. The signal loss dy is proportional to the signal value y and the number of the produced color centers, which are proportional to the absorbed dose dR . Crystal recovery is proportional to a difference between the asymptotic value y_0 (after recovery) and the current signal value. Also it is proportional to the recovery time dt :

$$dy = -P1 \cdot y dR + P2(y_0 - y)dt = -(P1 \frac{dR}{dt} + P2)y + P2 \cdot y_0 dt \quad (1)$$

In our case the dose rate($\frac{dR}{dt}$) was almost the same during the 85 hours of irradiation. Integration of this equation gives us the expression:

$$y = P0 \cdot \exp^{-(P1 \frac{dR}{dt} + P2) \cdot t} + \frac{P2 \cdot y_0}{P1 \frac{dR}{dt} + P2} \quad (2)$$

We can present the signal loss behaviour function as

$$f(t) = a \cdot \exp^{-t/\tau} + (1 - a), \quad (3)$$

The results of the fit for Fig. 11(a) are listed in Table II.

TABLE II. Results of fits to $f(t) = a \cdot \exp^{-t/\tau} + (1 - a)$

Signal Source	a	$\tau, hour$
Electron beam	0.104 ± 0.002	30 ± 2
Blue LED	0.054 ± 0.002	34 ± 5

The parameter a defines the saturated light loss value that is reached as t goes to infinity at a constant dose rate. Close to the asymptotic value, the crystal lost 10% in the electron signal and 5% in the blue LED signal. The τ parameter defines the saturation time constant, which is 30 hours for our crystal and our dose rate.

The time constants for the ten studied crystals are between 20 and 30 hours. There is no significant difference in τ for the LED and electron signals.

We should make a note at the end of this section. When a crystal is irradiated, the red LED light is slightly absorbed. Herewith, the blue LED light is absorbed more, in 3-6 times more [12] compare to the red LED light in the crystals. We can estimate that the electron signal is absorbed in about two times more than the blue LED signal (see Fig. 11(c)). We assumed that red light was unchanged under crystal irradiation, and assigned the PMT gain change to the red LED change. It means that the absolute electron signal loss values might be in about 1.1 times higher than the presented ones.

D. Irradiation by high-energy pions

After the electron irradiation program was finished, we irradiated the same crystals with pions for a four day period. We used a 40 GeV π^- beam. The size of the 40 GeV pion beam was 8 cm horizontally and 6 cm vertically, *i.e.* 90% of the beam was contained within these dimensions. The beam intensity was 6×10^6 /sec. Six crystals were irradiated with a dose rate ranging from 10 to 30 rad/h. Five cycles of irradiation (15-20 hours each) were alternated by low intensity electron beam exposures to measure the scintillation signals in the crystals.

The radiation damage region in the crystals is different for an electron and a pion irradiation (see Fig. 1). Thus, if a crystal was irradiated first by electrons until saturation in radiation damage was reached for a given dose rate, then we expect to get an additional signal loss with pion irradiation even at the same dose rate. Fig. 13 shows the additional loss of signal for one of the crystals (from Apatity). This crystal was irradiated by 27 GeV electrons and then by 40 GeV pions. During the 85 hours of e^- irradiation the dose rate was 12 rad/h. Then in the next 85 hours the dose rate was an order of magnitude less, and the crystal recovered. As a result, the first filled square point for pion irradiation data is above many open points for electrons. After that the crystal was irradiated by pions with the dose rate 12 rad/h for 100 hours. We see that the crystal lost 8% of the signal during the electron irradiation period and 14% of the signal during the pion

irradiation period with the same dose rate. The constant of proportionality between the blue LED and the electron signal is 0.3 for electron irradiation (if one fits by a straight line). This is about the same for pions at the beginning of the pion irradiation, but then increases up to 1 during the further pion irradiation. The crystals (manufactured in Bogoroditsk and Apatity) lost about 14% on pion irradiation. The surrounding crystals which were irradiated with a dose rate of about 1 rad/h lost less than 1 % of their light output.

The dependence of a signal loss on dose rate was studied in a separate run using 40 GeV pion irradiation. Each beam exposure lasted for 6 continuous hours. The beam intensity started from 2×10^5 /sec and was increased in a few steps up to 8×10^6 /sec by the end of the study. The beam was present in 1 sec of the full accelerator cycle of 9 sec. After each 6 hour irradiation exposure we lowered intensity by a few orders of magnitude, down to 3×10^4 /sec, so that we could avoid pile-up and see a minimum ionizing peak (MIP) for pions traversing the crystals without interacting. The crystals light output signals were monitored using the MIP peak; this procedure took 2 hours at low intensity. After that we took again high intensity beam exposure for the next 6 hours to continue irradiating the crystals. Then again switched to the low intensity MIP exposure.

To check our procedure for obtaining the change in scintillation light from time to time, we used pure muon beams and 27 GeV electrons to measure the light output changes due to pion irradiation. We continued this procedure of alternating high intensity and low intensity beams for 10 days in a row. The dependence of the normalized MIP signal on an absorbed dose for the two crystals in the array is shown in Figs. 14(a) and (b). (The normalized MIP signal is defined as the ratio of the MIP signal after some absorbed dose to the one before the pion irradiation).

We have observed the dependence of light output loss on the dose rate. Like electron radiation, the light loss exhibits saturation effect when the dose was kept at a constant level. The correlation between a change in the LED signal and a change in the MIP signal under irradiation was also measured (see Fig. 14(c) as an example). The constant of proportionality, if one fits by a straight line is different for different crystals and is on average 0.7 (the LED signal decreases less than the MIP one). In Fig. 15(a) we show the decrease in the LED signal for moderate dose rates. Different crystals received different absorbed doses during 10-day irradiation period. The open circles stand for Bogoroditsk crystals and the filled circles stand for Shanghai crystals. Six points in Fig. 15(a) represent the six crystals described above which accumulated absorbed doses of more than 1 krad each. Fifteen other crystals were irradiated by the beam halo and received absorbed doses less than 500 rad each. They are shown on the left side of Fig. 15(a). Three Bogoroditsk crystals are close to each other in their radiation hardness, however the twelve Shanghai crystals differ among each other by an order of magnitude.

Irradiation of lead tungstate crystals creates color centers which reduce the light attenuation length. One expects that the change of attenuation length will affect the longitudinal uniformity. This can degrade the energy resolution. On the other hand, if the loss of light collected in the crystal after irradiation is relatively small, the energy resolution itself might not be degraded so that the radiation damage can be regarded as only a calibration issue. The non-uniformity of the light yield (LY) along the crystal contributes to the energy resolution. To measure changes in the LY non-uniformity the crystal array was rotated by 90° with respect to the beam direction, before the irradiation by pions and just after the 10 days of irradiation. The crystals were scanned with the muon beam. The position of the muon track going through the crystal was reconstructed with the drift chambers. The data were binned along the crystal lengths in 1 cm intervals. The energy deposit distribution was fitted in each bin by a convolution of Gaussian and Landau function. The non-uniformity of the light yield in the front part of crystal (3-10 radiation lengths) was about 0.5 %/cm. The non-uniformity did not change significantly after a dose up to 4 krad at a dose rate of up to 60 rad/h, which caused the signal loss of up to 30%. As a result, the energy resolution of the crystals did not change. The relation between the change in transparency seen by the LED light and the change seen by the scintillation light varies from crystal to crystal. A plot of such constants of proportionality for seven crystals is shown in the Fig. 15(b). The first four points show the Shanghai crystals, and the next three points show the Bogoroditsk crystals. Points 4 and 5 represent the super-intensive dose rates obtained by the Shanghai crystal S25 and the Bogoroditsk crystal B21 (details will be given in the next Section). We can see that the constants of proportionality for 40 GeV pion irradiation (Fig. 15(b)) are larger than the constants of proportionality for 27 GeV electron irradiation (Fig. 11(c)).

After the irradiation by electrons and then pions was finished, we kept the PMT HV on and studied crystal recovery for 15 days using the LED pulser. The results for the Apatity 1434 crystal are presented in Fig. 16. We fitted the dependences of recovery on time for the six crystals with an exponential function. The average recovery time is (200 ± 40) hours, and the LED damage recovery for 400 hours is $(87\pm 5)\%$ for these six crystals.

IV. SUPER-INTENSIVE BEAM IRRADIATION

Six crystals from Bogoroditsk and Shanghai were irradiated by secondary particles coming out the internal target of the 27-th magnet block of the Protvino U-70 accelerator(see Fig.17). Two of them were irradiated at a dose rate of 100 krad/h, and the other four at 1 krad/h. For the latter case the intensity of the primary proton beam was lowered by two orders of magnitude. To measure the absorbed dose, thermo-luminescence dosimeters (TLD) were attached to the front face of the crystals. They were of LiF type doped by Mg, Cu and P, 5 mm in diameter and 200 μm in thickness. In addition, an ionization chamber(IC) filled by Xenon was installed behind the crystals. The sensitive volume of the chamber was as 18.5 mm in diameter and 36 mm in length. Both TLDs and the IC were calibrated using a Cs-137 gamma source. The accuracy of the absorbed dose measurements by TLDs and IC in this mixed radiation field was estimated to be 30% each. These measurements were in general agreement with the results of the MARS calculations; the worst case difference was a factor of 1.5. The dominant systematic error of the calculations was due to the accuracy of the irradiation facility geometry. The IC was used to monitor the number of protons produced at the internal target for each run of the crystal irradiation. *Al* activation detector in Fig.17 was used to measure a fluence of the hadrons (number of hadrons per cm^2) with energy greater than 20 MeV.

Two crystals, Bogoroditsk B21 and Shanghai S25, were irradiated in the first exposure at about 100 krad/h dose rate. The longitudinal profiles of the absorbed dose rates are shown in Fig. 3(a). The maximal values of the absorbed doses accumulated in crystals during the five exposures are given in Table III. Four crystals, Bogoroditsk B17, B9 and Shanghai S22, S18, were irradiated in the second exposure at 1 krad/h dose rate. The longitudinal profiles of the absorbed dose rates are shown in Fig. 3(b). The absorbed doses accumulated in the crystals during the five exposures are given in Table IV.

TABLE III. The maximal values of the absorbed doses accumulated in Bogoroditsk B21 and Shanghai S25 crystals during the five exposures at the IHEP irradiation facility

Exposure (minutes)	Bogoroditsk B21 (krad)	Shanghai S25 (krad)
0.83	3.4	1.8
9.67	40	21
66	270	140
475	1970	1020
747	3100	1610

The results of the irradiation of the two crystals in the first exposure are presented in Fig. 18. The procedure was to irradiate the crystals and then measure their light output immediately thereafter using the 27 GeV electron beam. In some cases, we measured the light output again after letting the crystals sit without any radiation. The Bogoroditsk crystal (see Fig. 18(a)) lost 33% of the initial signal after first 3.4 krad dose. After the second irradiation, the absorbed dose increased up to 43 krad and the signal loss was increased up to 46%. After 47 hours of recovery time, the signal rose up to 70%. After the third irradiation, the Bogoroditsk crystal accumulated 313 krad and the signal was at the level of 49%. 32 hours of recovery time returned it to a level of 57%. After the fourth dose the total radiation was 2300 krad and the signal level

TABLE IV. The maximal values of the absorbed doses accumulated in Bogoroditsk B17, B9 and Shanghai S22, S18 crystals during the four exposures at the IHEP irradiation facility

Exposure (minutes)	Bogor. B17 (krad)	Bogor. B9 (krad)	Shanghai S22 (krad)	Shanghai S18 (krad)
25	0.7	0.7	0.35	0.35
72	2	2	1	1
60	1.7	1.7	0.8	0.8
60	1.7	1.7	0.8	0.8

was at 37%. After 15 hours recovery time, the signal recovered slightly to 39%.

The Shanghai crystal (see Fig. 18(b)) lost 18% of the signal after first 1.8 krad dose. After the second irradiation, the integrated dose increased up to 23 krad and the signal loss increased up to 33% relative to the signal before the irradiation. After 47 hours of recovery time, the signal rised up to 69%. After the third irradiation, the Shanghai crystal accumulated 163 krad and the signal was at the level of 66%. One should mention that the signal was pretty stable between the second and the third irradiations including recovery time and was at the level 66-69% for the absorbed doses of 23-163 krad. After getting 2800 krad, the signal dropped down to 33%.

One of the most important conclusions of this work is that even after an integrated dose about 2.5 Mrad obtained with a super-intensive dose rate 100 krad/h both crystals remained usable, although they lost 2/3 of their light. In BTeV we expect that only 0.1% of the crystals will receive this much dose in a year. As was expected, the constants of proportionality in the MIP-Electron correlations for both the crystals are about 1. The LED-Electron correlations for both the crystals are shown in Fig. 19. The constants of proportionality for the two crystals are 0.5 and 0.66. The degradation of single crystal energy resolution for 27 GeV electrons was only 20% for Bogoroditsk crystal and 50% for the Shanghai crystal.

The four crystals irradiated in the second exposure with a dose rate of 0.5-1 krad/h, and a total dose of 350-700 rad, lost up to 10% of their light output for Shanghai crystals and up to 25% for Bogoroditsk crystals. After each of the next three runs no signal loss was seen, within the 3% accuracy (the systematic error due to a PMT gain change effect).

V. SUMMARY AND CONCLUSIONS

Radiation hardness of lead tungstate crystals is an important issue for the BTeV experiment at Fermilab. Simulation of absorbed dose profiles in the crystals with the use of the MARS program has shown that the dose rates for the crystals range from 0.1 up to 700 rad/h. About 95% of the crystals in the BTeV electromagnetic calorimeter will get the absorbed doses from 0.1 to 30 rad/h assuming that the Tevatron luminosity is $2 \times 10^{32} cm^{-2} sec^{-1}$. Almost 5% of the crystals will get from 30 rad/h up to 200 rad/h, and only 0.5% more than 200 rad/h.

A study of radiation damage in lead tungstate crystals has been carried out in Protvino in 2001-2002 for BTeV. The crystals were manufactured in Bogoroditsk (Russia) and Shanghai (China) at the very end of 2000, and in Apatity (Russia) in early 2002. There were two approaches in the study. First, crystals were irradiated by high-intensity high-energy electron and hadron beams at radiation doses ranging from 0.1 to 60 rad/h. Secondly, crystals were irradiated by charged hadrons, γ -quanta and neutrons from the internal target of the U70 in a wide energy spectrum from 10 eV up to 70 GeV at dose rates between 0.5 and 100 krad/h.

The dependence of light output loss on a dose rate has been measured. The light loss exhibited saturation when the dose rate was kept constant. At larger dose rates, the light output loss still saturates but at lower light output levels. Each crystal had a different percentage of light loss when it saturated. More quantitatively: no light output loss was observed for dose rates less than 1 rad/h. For dose rates of 10-25 rad/h with 27-GeV electron irradiation, eight crystals lost on the average 8%. For 40 GeV pions this average was 12% at comparable irradiation dose rates. The difference between the damage due to electron and pion irradiation can be attributed entirely to their difference in the radiation profile along the length of the crystal. Much of electron energy is deposited near the shower maximum, from 4 to 10 cm from the front of the crystal. For pion beams, the radiation dose profile reaches its maximum around 5-7 cm and stretches all the way to the rear-end of the crystal. However, a possible effect due to the difference between the physical processes by which electrons and pions interact with crystals cannot be ruled out.

For dose rates of 30-60 rad/h using 40-GeV pion irradiation, five crystals lost on the average 20%. For a dose rate of 500 rad/h using irradiation by charged hadrons, γ -quanta and neutrons with the average energy of 10 GeV, two crystals lost 10%, and two other crystals lost 25% when they were exposed to 1 krad/h of radiation. Two crystals got extremely high dose rate of 100 krad/h and accumulated about 2.5 Mrad absorbed dose (maximum annual dose of any BTeV crystals!) also with the same mixed particle spectra irradiation. They remained useable. Their light output loss was a factor of 3. This is far from the BTeV environment, where 700 rad/h will be the highest 0.1% crystals.

There is a correlation between a change in the LED signal and a change in the beam (electron or MIP) signal under irradiation. The constant of proportionality is different for different crystals and varies from 0.3 to 0.6 for electron irradiation and from 0.5 to 0.9 for pion irradiation.

The non-uniformity (maximum 0.5% per cm at one third of the crystal length) of the light yield does not change significantly when the dose rate is up to 60 rad/h. After 2.5 Mrad absorbed dose with a dose rate of 100 krad/h the uniformity became 1.5 times poorer, at least for one of our crystals.

When irradiation decreases or stops, crystals recover. The average recovery time for six crystals which lost from 7 to 20% of the LED signal, is (200 ± 40) hours, and the damage recovery after 400 hours was $(87 \pm 5)\%$.

To summarize, lead tungstate crystals lose light from irradiation by high-intensity high-energy beams. This loss level depends on dose rate. If dose rate does not change, the light loss saturates. If the dose rate is reduced, the light output recovers. Crystals have to be calibrated continuously during the BTeV experiment. We did not see a significant difference in radiation hardness of the crystals from the three manufacturers.

VI. ACKNOWLEDGMENTS

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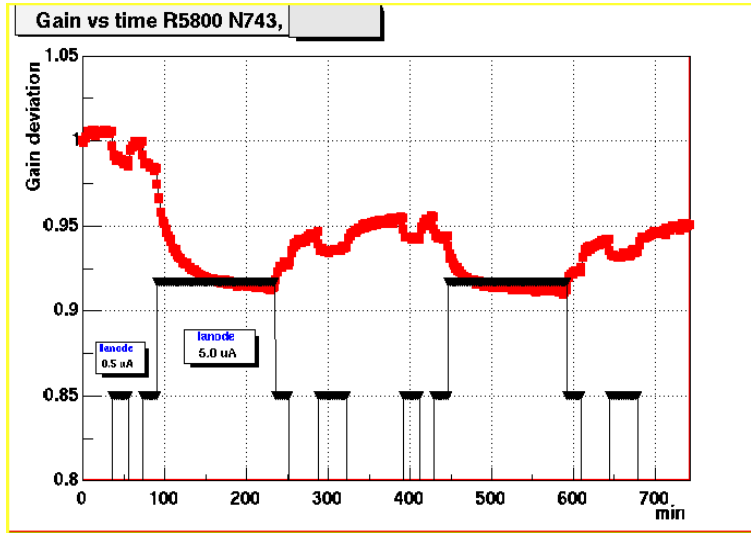


FIG. 8. The behavior of PMT number 743 at the dedicated stand for PMT gain variation measurements. (See text for details.)

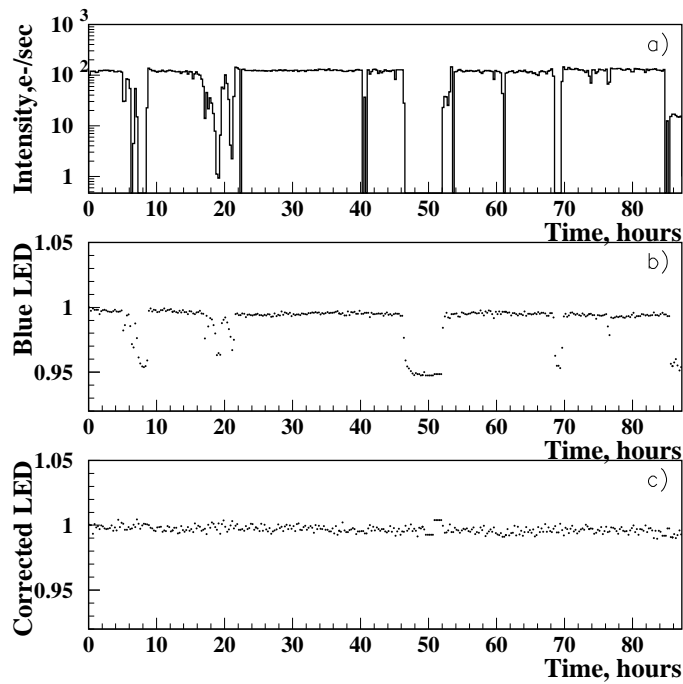


FIG. 9. (a) Electron beam intensity in the Shanghai crystal S22 over time. Blue LED time behavior in this crystal (b) before and (c) after correction using red LED data.

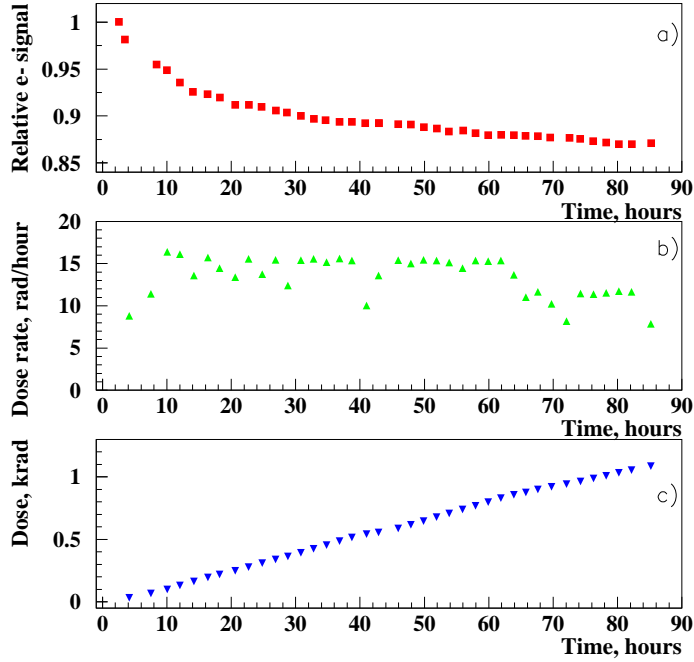


FIG. 10. (a) Normalized electron signal during 85 hours of irradiation by 27 GeV electrons for the Bogoroditsk crystal B14. (b) Electron beam intensity in dose rate units. (c) Absorbed dose.

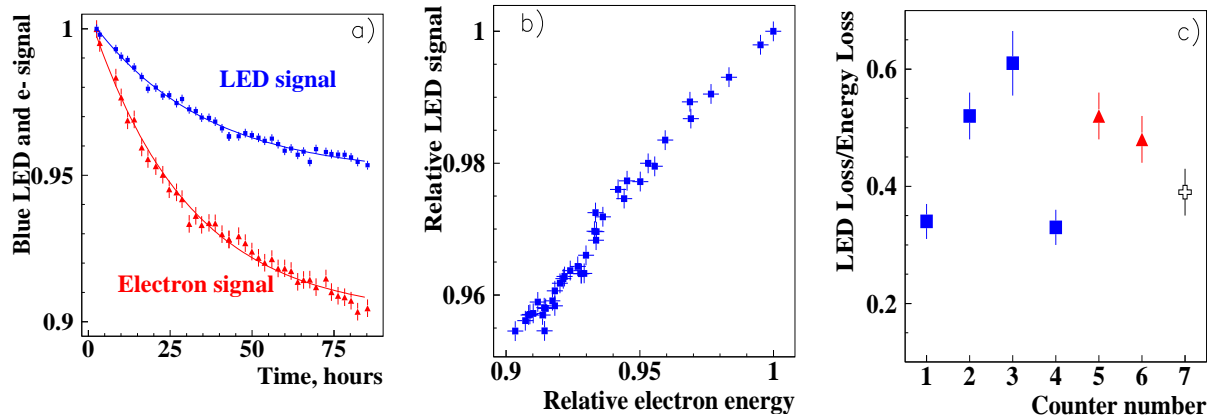


FIG. 11. (a) Blue LED and electron signals for the Shanghai crystal S22, which was irradiated by 27 GeV electrons with a dose rate of 16 rad/h. (b) Blue LED-electron correlation for the same crystal. (c) LED-electron correlation coefficients for the seven crystals. Irradiation was by 27 GeV electrons. Square points stand for the Bogoroditsk crystals B12, B13, B14 and B17, triangular points stand for the Shanghai crystals S14 and S22, and a crest point stands for the Apatity crystal 1447.

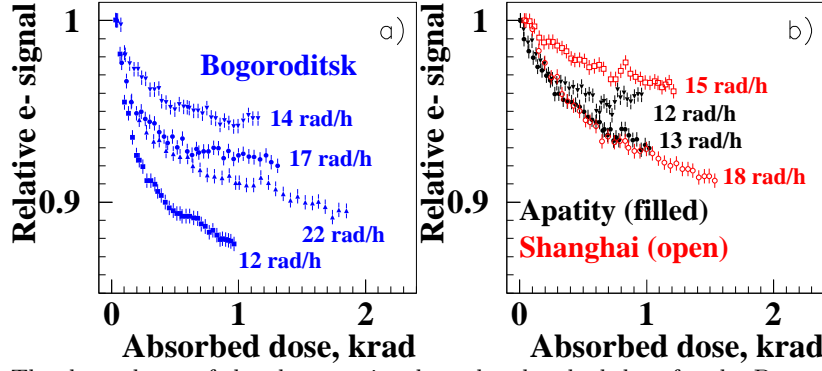


FIG. 12. (a) The dependence of the electron signal on the absorbed dose for the Bogoroditsk crystals B14, B22, B12, B16, (b) for the Shanghai (open points) S22, S14 and the Apatity (filled points) 1447, 1434 crystals. Each crystal was irradiated by 27 GeV electrons at the fixed dose rate (different for each crystal) for 85 hours.

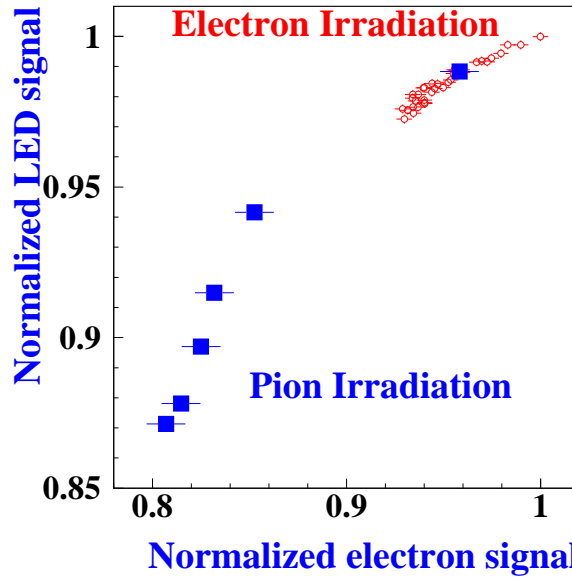


FIG. 13. Normalized LED signal versus normalized electron signal for Apatity crystal 1447. Open points show electron irradiation and filled square points show further pion irradiation.

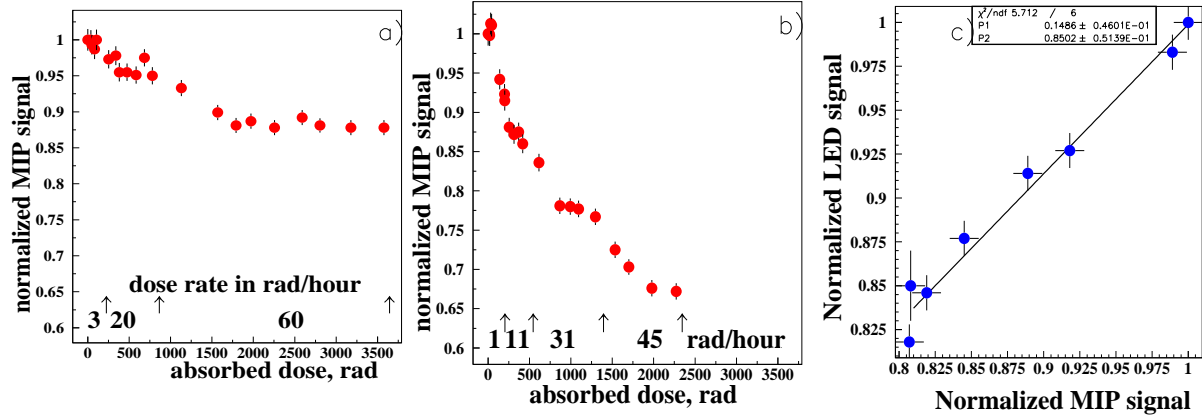


FIG. 14. Dependence of the normalized MIP signal on absorbed dose for (a) Shanghai crystals S16 and (b) S20. (c) Correlation between the LED and the MIP signals for Shanghai crystal S19 as a result of a pion irradiation.

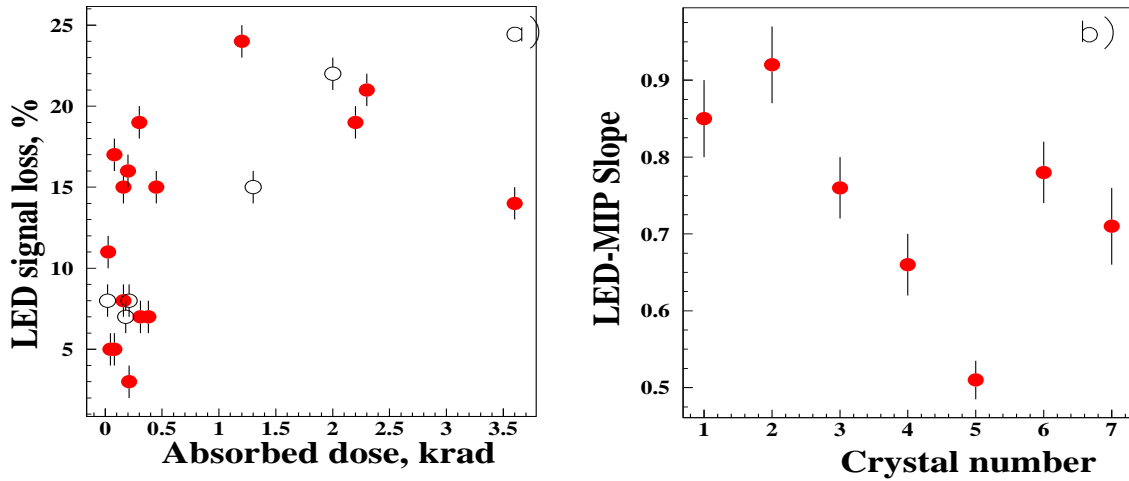


FIG. 15. (a) Dependence of a LED signal loss on absorbed doses obtained for a 10 day irradiation by a 40 GeV pion beam of the 21 crystals. (100 rad = 1 Gy). (b) The constants of proportionality between the change observed by the LED monitoring system and the change in the beam (MIP) signal for seven crystals. Irradiation was by 40 GeV pions.

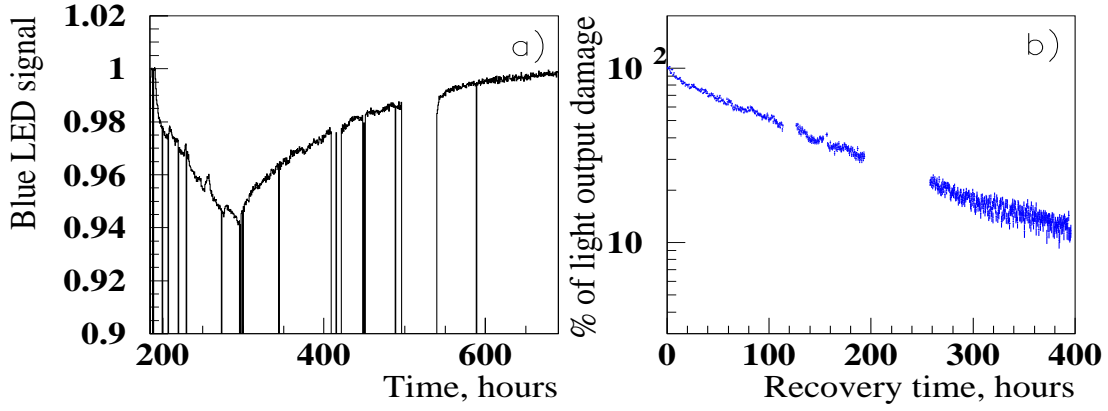


FIG. 16. (a) Blue LED signal of the Apatity 1434 crystal which was irradiated by 40 GeV pions during 100 hours and then recovered during the next 400 hours. (b) The blue LED light output damage recovery in the same crystal. We see that the crystal recovered 90% of its light output damage after 400 hours.

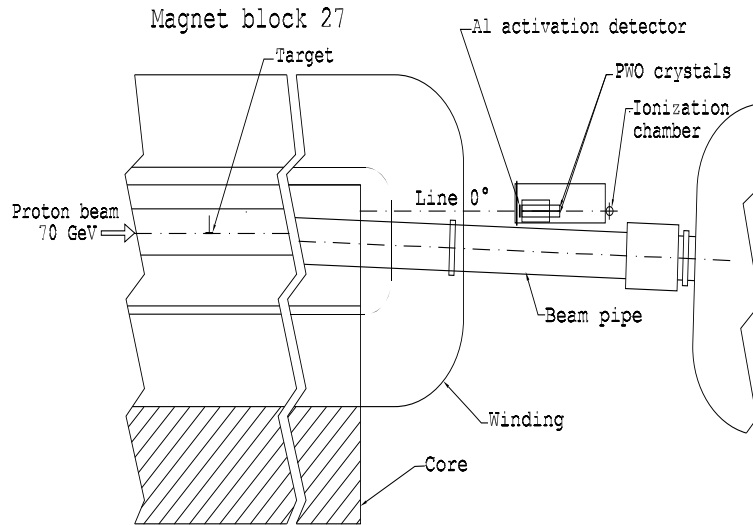


FIG. 17. Superintensive dose irradiation facility

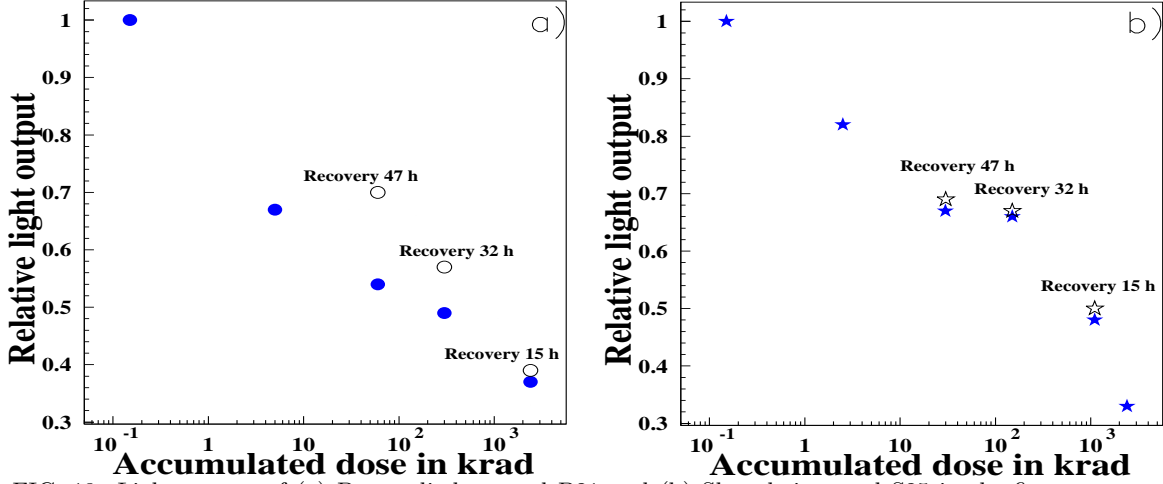


FIG. 18. Light output of (a) Bogoroditsk crystal B21 and (b) Shanghai crystal S25 in the first exposure versus absorbed dose in krad at the 27 GeV electron beam after different steps of irradiation procedure. Low points at each absorbed dose stand for light output just after the irradiation. Upper points (if any exist) stands for light output after some recovery time.

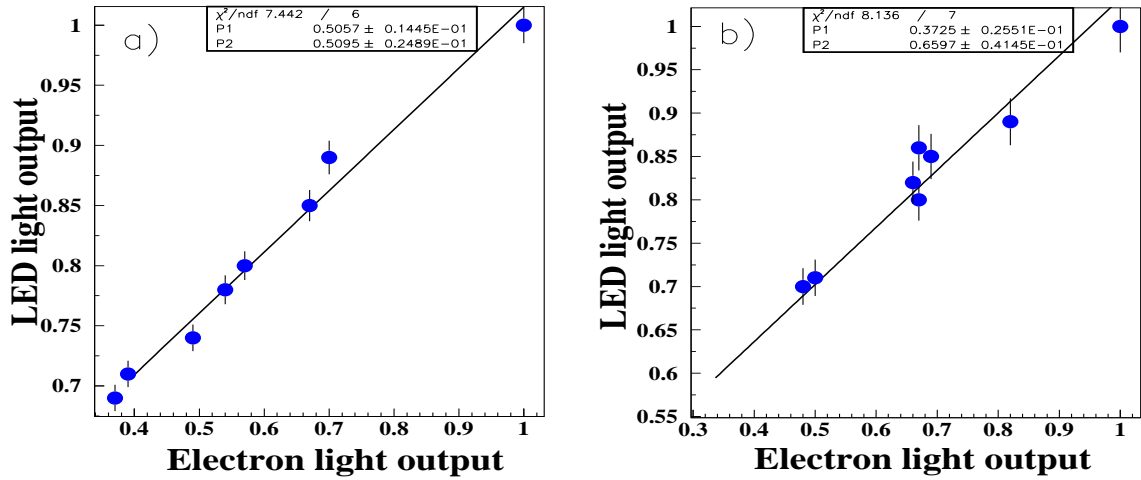


FIG. 19. Correlation between the LED and the beam electron signals for (a) Bogoroditsk crystal B21 and (b) Shanghai crystal S25.