Nuclear Instruments and Methods in Physics Research A234 (1985) 435–442
North-Holland, Amsterdam

RADIATION DAMAGE OF BGO AND CsI(Tl) CRYSTALS

Ch. BIELER, D. BURKART, J. MARKS, M. RIEBESELL, H. SPITZER and K. WITTENBURG

II. Institut für Experimentalphysik der Universität Hamburg, Fed. Rep. Germany

G.-G. WINTER

Deutsches Elektronen-Synchrotron DESY, Hamburg, Fed. Rep. Germany

Received 29 May 1984

We have measured the response of five 10–20 cm long BGO crystals from different manufacturers to irradiation with ^{137}Cs γ-rays at doses of 40 and 85 rad. Immediately after irradiation the pulse height drops by 26–38% and recovers only partially with time. 110 d after irradiation the remaining damage is between 1 and 13%. A 10 cm long CsI (Tl) crystal shows pulse height reductions after irradiation which do not recover with time. The cumulative effect of a continuous irradiation on a BGO crystal ($1 \times 1 \times 15$ cm^3) in a partially shielded position on the beam pipe of PETRA was measured over a period of 53 d. At an average daily dose of 1.9 rad the pulse height dropped continuously resulting in an overall pulse height loss of 9% in 7 weeks. This indicates that BGO when applied in long and narrow shapes is more sensitive to small radiation doses than previously assumed.

1. Introduction

Recently several projects have been put forward which propose the use of bismuth-germanate (BGO) as the scintillator material of compact electromagnetic calorimeters at e^+e^- storage rings [1–4]. One of the properties which might limit the use of BGO ($Bi_4Ge_3O_{12}$) at storage rings is its sensitivity to damage by synchrotron and electron background radiation. Typical radiation levels to be expected are e.g. 100–2000 rad/month on the beam pipe of PETRA and 30–100 rad/month at a radial distance of 25 cm from the interaction point of the DORIS ring.

Initial work on the radiation resistance of short BGO crystals (length $\lesssim 3$ cm) concentrated on high dose exposures between 10^3 and 10^6 rad [5,6]. Recently the response of longer crystals with lengths between 10 and 20 cm to doses between 1 and 4000 rad has been studied [2,7,8]. From these studies the following picture emerges:

1) After a short term exposure the pulse height of BGO decreases substantially (up to 40%).
2) At low doses (1–4 rad) the initial damage grows linearly with the dose [2], whereas at larger doses (above 100 rad) saturation effects occur [8].
3) The induced damage recovers spontaneously with at least three different time constants, the longest being on the order of 3 weeks [8].
4) The radiation damage and recovery rates differ from crystal to crystal [8].

The authors have given substantially differing estimates on the tolerable continuous dose which causes an asymptotic pulse height loss of less than 1%. Estimates vary between 0.9–3.8 rad/d [2] and 2000 rad/d [6].

We have thus concentrated on measuring the response of BGO to low doses (below 100 rad) of γ-radiation. We have chosen two approaches. Firstly, we have measured the long term response to short exposures. We find that some crystals suffer a pulse height loss from which they do not recover on a time scale of 100 d. Secondly, we have exposed a partially shielded crystal to the radiation near the beam pipe of the e^+e^- storage ring PETRA over an extended period of time. We find a cumulative effect which indicates that long BGO crystals are more sensitive to small radiation doses than previously assumed. Recently CsI(Tl) has been discussed as an alternative to BGO. We therefore have performed a radiation test on a CsI crystal ($1.5 \times 1.5 \times 10$ cm^3) with doses of 42 and 85 rad.

2. Short-term exposures

2.1. Experimental procedure

In the following we first describe the setup for pulse height measurements and subsequently the irradiation procedure. Six BGO crystals and one CsI(Tl) crystal have been tested. The properties of the crystals are listed in table 1. The crystals have all sides polished. The Harshaw crystal is slightly yellow. One end of the Crismatec crystal is intensely yellow. The remaining crystals are clear and colorless. All crystals were wrapped

0168-9002/85/$03.30 © Elsevier Science Publishers B.V.
(North-Holland Physics Publishing Division)

Table 1
Properties of the test crystals

Crystal	Supplier	Size (mm^3)	Growing method	Delivery date	Cs res. $\Delta E/E$ [a]	Uniformity [b]
BGO	Harshaw Holland [9]	15×15×200	Czochralski	Jul. 83	24.7%	20%
BGO	Crismatec [10]	15×15×200	Czochralski	Aug. 83	23.5%	16%
BGO	NKK [11]	11×11×200	Czochralski	Feb. 83	19.3%	17%
BGO	NKK	10×10×20	Bridgeman	Dec. 82	16.6%	–
BGO	Inst. of Ceramics Shanghai [12]	15×15×200	Bridgeman	Jul. 83	18.7%	12%
BGO	Scholz [13]	15×15×100	Czochralski	Aug. 83	17.3%	2%
CsI	Korth [14]	15×15×100		Oct. 83	9.8%	13%

[a] Head on energy resolution of ^{137}Cs γ-rays ($E_\gamma = 662$ keV). ΔE is full width at half height.
[b] Uniformity of pulse height (ph) given by (max. ph − min.ph)/(max. ph) measured over 16 cm (8 cm) of the crystal length for 20 cm (10 cm) long crystals.

in 50 μm thick white reflective foil (Dupont, Tedlar 200 BS 30 WH). The crystals were coupled with optical grease (Rodorsil huile 47 V) to Hamamatsu R268 photomultipliers, which have a spectral response from 300–650 nm with a maximum at 420 nm. During handling the crystals were kept under red light.

The readout setup is shown in fig. 1. The photomultiplier was connected via a charge sensitive preamplifier (Canberra 2005) and a shaping amplifier (Ortec model 451, shaping time 1 μs) to a multi-channel-analyzer (LeCroy 3001). The drift of the multipliers and amplifiers was monitored by LED pulses. The LEDs (CQY 66 from ITT) were glued to the photocathode window close to the crystal.

In order to minimize temperature variations each crystal was wrapped by a cover of 2 cm thick styrofoam as shown in fig. 2. The crystal temperature was monitored by a temperature sensor mounted near the crystal. Fig. 3 shows the temperature variation during the measuring period. The average temperature was 296 K. The results given below have been corrected assuming a temperature coefficient of −1.5% K^{-1} for BGO and −0.3% K^{-1} for CsI. The scintillation light yield was measured by positioning an uncollimated 10 μCi ^{137}Cs γ-source 5 cm away from the narrow end face of the crystal. Table 1 shows the resulting energy resolution (fwhm). The longitudinal uniformity was measured by moving a collimated ^{137}Cs source along the crystal. Results are also shown in table 1.

The irradiation was performed by exposing the crystals to a strong ^{137}Cs source available at DESY. The crystals were decoupled from the multipliers and irradiated uniformly over the full length. The radiation dose was measured by thermoluminescent dosimeters (TLD) mounted on the side of the crystal facing the source and on the opposite side. Because of attenuation the detected doses on the rear side are less than half as large as on the front side *. Table 2 shows the dose values. Two irradiations were performed. The first lasted 18 h with an integrated dose of 85 rad, the second one yielded 40

* The dose values quoted below give the dose absorbed by the TLDs on the front side. Note that the absorption coefficient for TLDs is 1.16 times higher than for air. We understand that the dose values quoted in the references refer to doses absorbed in air equivalent.

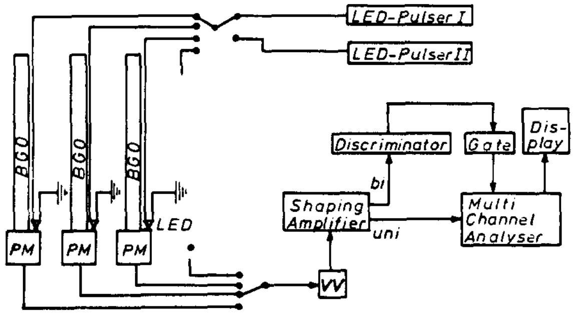

Fig. 1. Readout setup for measuring pulse heights from BGO and CsI scintillating crystals.

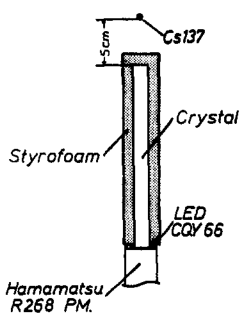

Fig. 2. Arrangement of crystal and photomultiplier.

Table 2
Radiation doses and exposure times of two irradiations by a ^{137}Cs γ-source

	Crystal (size (mm^3))	TLD doses (rad)		Exposure time (h)
		front face	opposite face	
(a) first exposure	Harshaw (15×15×200)	85.5 87.8 82.9	30.7 30.6 33.8	18
	Crismatec (15×15×200)	84.4	–	18
	NKK (11×11×200)	87.5 85.3 83.3	– –	18
	Shanghai (15×15×200)	87.5 85.3 83.3	35.4 35.4 34.8	18
	Scholz (15×15×100)	84.0 84.9 81.4	31.4 34.7 32.8	18
	NKK (10×10×20)	39.4	–	24
	CsI(Tl) (15×15×100)	41.7±10	–	24
(b) second exposure	Harshaw (15×15×200)	40	–	24
	Crismatec (15×15×200)	42	–	24
	NKK (11×11×200)	37	–	24
	CsI(Tl) (15×15×100)	85	–	24

rad in 24 h. The values for CsI were 42 rad and 85 rad in 24 h each, implying instantaneous doses of the order 1.75–3.5 rad/h. The uncertainty of the dose measurement is typically 10%. After irradiation the crystals were coupled to multipliers again in a well defined position. The systematic uncertainty of the pulse height measurement is ±1.5%. The first measured point after irradiation has a larger uncertainty of ±5% due to turn on effects of the photomultiplier. Measurements started about 15 min after the end of a irradiation. Hence short term pulse height changes during and immediately after irradiation are not monitored.

2.2. Results of the exposures of BGO

In the following we present the results of the pulse height measurements after the exposures. The overall time monitored for most of the crystals is 180 d. Figs. 4(a)–(f) show the relative pulse height where 100% corresponds to the pulse height before the first irradiation.

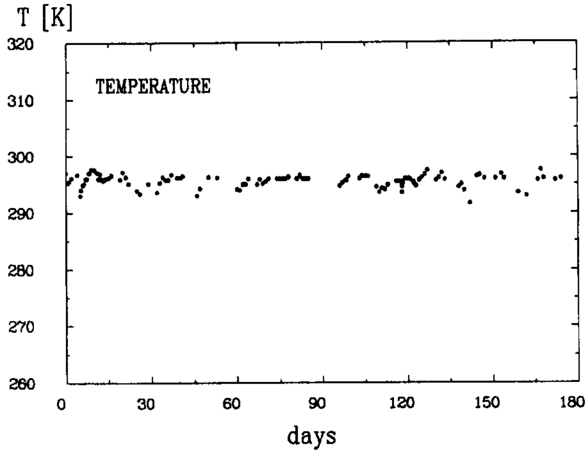

Fig. 3. Temperature history of the crystals used for short term radiation exposures to a ^{137}Cs γ-ray source.

We observe the following behavior (see table 3):
1) After the first exposure of about 85 rad the pulse height drops by up to 40%.
2) All crystals show some degree of spontaneous recovery.
3) The recovery pattern of the long crystals (figs. 4(a)–(c), (e), (f)) is consistent with three time constants as observed by Bobbink et al. [8].
4) In contrast to their conclusion the recovery is only partial. After about 40 d a permanent damage between 1% (Crismatec) and 13% (Scholz) remains.
5) Both the initial and the permanent pulse height reductions vary strongly from crystal to crystal.
6) The short (2 cm long) NKK crystal (fig. 4(d)) recovers very fast i.e. within one day and shows no permanent damage. This indicates that the long recovery times and the permanent pulse height losses observed in the longer crystals are mainly caused by damages to the transmission properties of the crystal [8].

After the second exposure of 40 rad we observe the following (see figs. 4(a)–(c)):
1) The recovery after the initial pulse height drop occurs faster, typically within ten days.
2) Whereas the Harshaw and NKK crystals suffer further permanent pulse height reductions, the Crismatec crystal reaches the original pulse height within errors.
3) When comparing the initial damage from the 40 and 85 rad exposures saturation effects seem to be present in the Harshaw and NKK crystals. Much stronger saturation effects have been observed by Bobbink et al. for doses above 100 rad.

Two of our results have not been reported before: (i) the occurrence of permanent damage at doses of 40–80

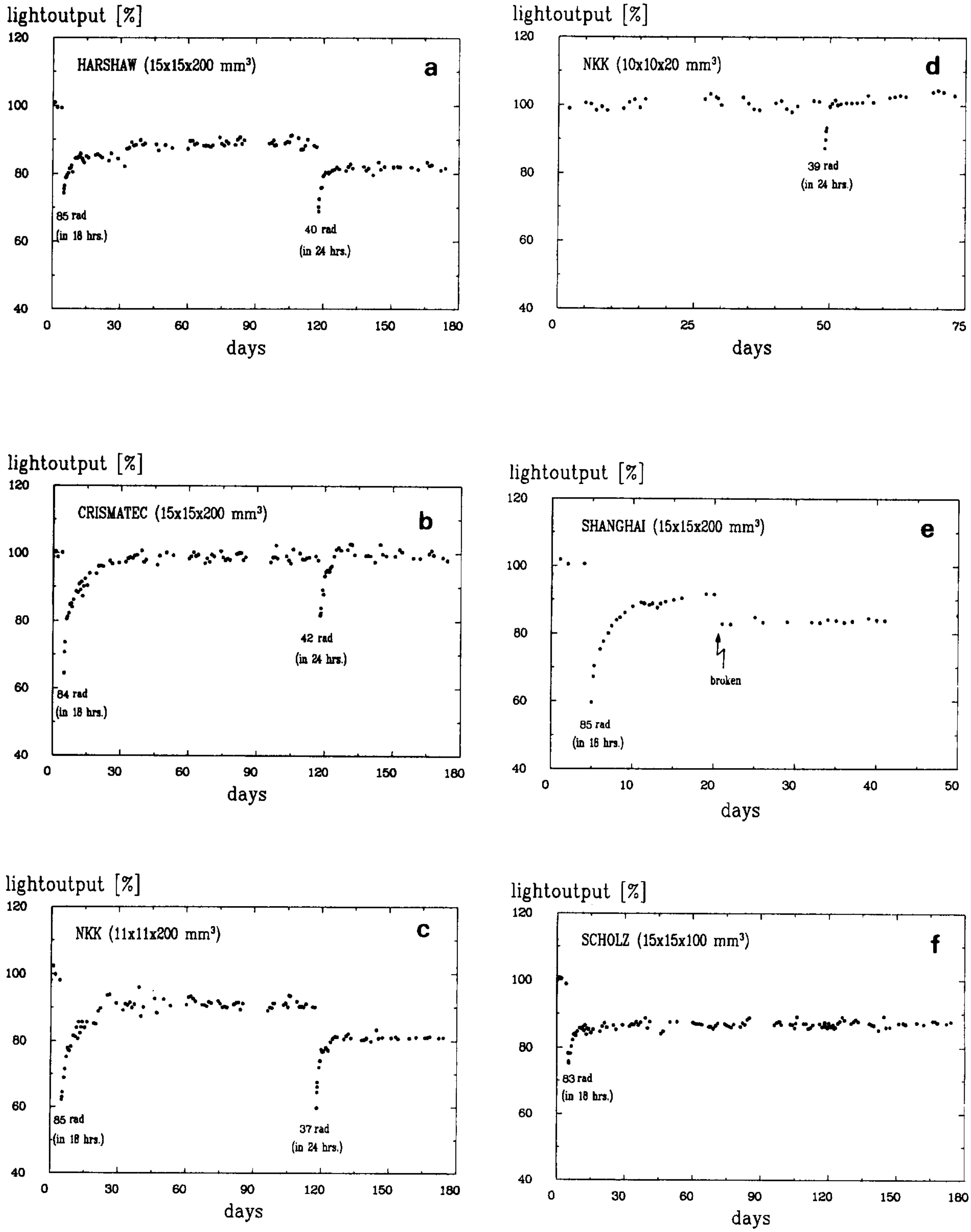

Fig. 4. (a)–(f) Pulse height history of various BGO crystals which were subject to short term radiation exposures. Plotted is the light output normalized to the light output measured before the first exposure. The radiation doses refer to the readings from the TLDs mounted on the front face of the crystal.

Table 3
Pulse height observed after two exposures (ph-100 before first exposure)

Crystal	First exposure (85 rad)		Second exposure (40 rad)	
	ph after (15 min)	ph after (100 d)	ph after (15 min)	ph after (30 d)
Harshaw (15×15×200)	74±4	89±0.5	70±3.5	82±0.5
Crismatec (15×15×200)	64±3.5	99±0.5	82±4	100±0.5
NKK (11×11×200)	62±3	91±0.5	60±3	81±0.5
Shanghai (15×15×200)	60±3	93±0.5 [a]	–	–
Scholz (15×15×100)	75±4	87±0.5	–	–

[a] The pulse height of the Shanghai crystal displays a drop after 20 d due an accident leading to a broken edge. The pulse height given in table 3 is an extrapolated value.

rad and (ii) the shorter recovery times present in the second exposure. The first result was probably missed by the authors of refs. [2,7,8] due to shorter observation times (at most 25 d between two exposures). Both results could be a special property of low dose irradiations. We cannot judge to what degree the results are caused by crystal impurities and damages in particular regions of the transmission spectrum. Note that the sensitivity maximum (420 mm) of the phototubes is below the emission maximum of BGO (480 mm). It is known from ref. [6] that reductions of transmission are strongest at small wavelengths.

Finally we would like to add that long term (non-self-recovering) damages like the ones observed by us can be annealed completely by heating the crystal up to 100°C [15].

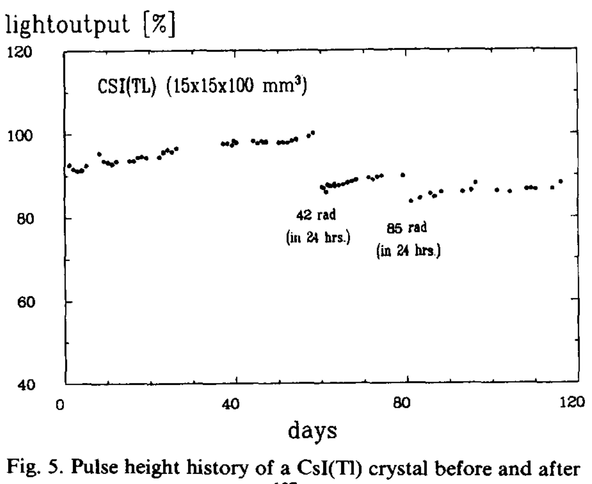

Fig. 5. Pulse height history of a CsI(Tl) crystal before and after two radiation exposures to a ^{137}Cs γ-ray source.

2.3. Results of the exposure of CsI (Tl)

Fig. 5 shows the pulse height history of the CsI crystal. The first irradiation was performed after 59 d of crystal monitoring, the second one after 21 further days. The pulse height measurement could only be resumed 16 h (!) after the end of irradiation, since a strong afterglow made the signal from the ^{137}Cs source undetectable. CsI (Tl) is known for its afterglow [16].

The figure shows three remarkable features:

1) Before the first irradiation the crystal displays a continuous pulse height rise of about 4%/month. This rise also seems to be present after the two irradiations. The rise could be due to a previous exposure to room light at the factory. The crystal was handled under red light at all times following delivery.
2) After irradiations of 42 and 85 rad, the pulse height drops by 13% and 7%, respectively; i.e. the damage saturates strongly at the second exposure.
3) Apart from the continuous pulse height rise of 4%/month we observe no long term recovery within errors of 2%. We cannot exclude the presence of additional fast-recovering damages (as known from NaI, see e.g. ref. [8]), which might occur on a time scale below 16 h.

In summary, the radiation damages in the CsI (Tl) crystal studied by us are of similar magnitude as the nonrecovering damages in BGO crystals. However, in view of the short length of our crystal (10 cm) and the longer radiation length of CsI ($X_0 = 1.86$ cm), we expect larger damages in full scale CsI calorimeters.

3. Long term exposure at PETRA

3.1. Experimental setup

A long term exposure of 53 d was performed at a partially shielded location on the PETRA beam pipe next to the CELLO detector. Fig. 6 shows a side view of the detector layout. A box containing the BGO crystal was put on the beampipe about 60 cm away from the mini β-quadrupole (l). The average radial distance of the crystal from the beam was 15 cm. The beampipe was wrapped between points (n) and (o) by 4 mm of lead. Further radiation protection was provided by a lead brick wall (k) total thickness 15 cm) and a lead glass shower counter (h). Three beam halo absorbers were placed in the beam pipe at positions (n), (o) and (p). The box itself was covered by a layer of 1 mm lead.

Fig. 7 shows the box containing a BGO crystal ($10 \times 10 \times 150$ mm^3) glued to a photomultiplier R 268. The crystal was delivered by Harshaw, Holland, in September 1982. It is a clear crystal with head-on resolution of $\Delta E/E = 17.3\%$ for ^{137}Cs γ-rays. A 10 μCi ^{137}Cs source was mounted inside the box 3 cm away from the crystal. The multiplier was readout in a similar way as described above. The drift of the system was monitored by a LED and a temperature sensor mounted inside the box. Continuous pulse height measurements were performed, even during injection.

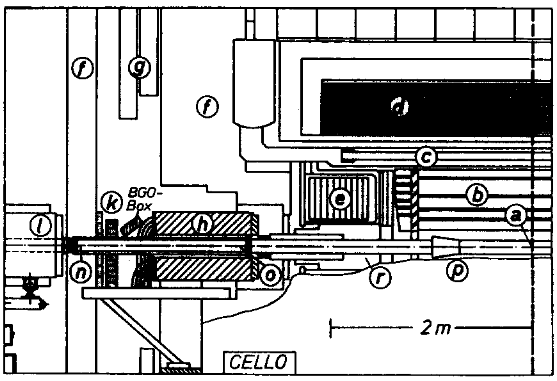

Fig. 6. Side view of the environment of the CELLO detector, where a BGO crystal was exposed to background radiation. The components relevant to the test are (a) interaction point, (c) solenoidal magnet coil, (e) endcap liquid argon counter, (f) magnet iron yoke and massive iron shielding wall, respectively, (h) lead glass shower counter (17 radiation lenghts), (k) two lead walls of total thickness 15 cm surrounding the vacuum pipe, (l) mini-β-quadrupole, (n), (o), (p) positions of beam halo absorbers (for details on the absorbers and further details of the radiation tests see ref. [17]).

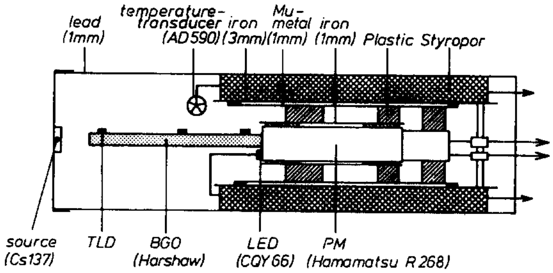

Fig. 7. Details of the BGO-box exposed to background radiation on the PETRA beampipe at the position shown in fig. 6.

3.2. Radiation dose and energy spectrum

Three TLDs were mounted on the side of the crystal facing the beam. The measured integrated doses were 119, 75 and 114 rad at three positions with an estimated average of 100 rad *. Although we did not measure the day to day dose variations, we expect that the total dose was not received in one or two big accidents but rather in a more continuous way. PETRA has typically five successful fillings per day. The exposure covered luminosity running at beam energies between 21.6 and 22.6 GeV.

In order to illustrate the energy range of the radiation hitting the crystal we show in figs. 8(a)–(c) three energy spectra measured (a) during injection ($E_{\text{beam}} =$ 6.9 GeV), (b) during a typical luminosity run ($E_{\text{beam}} =$ 23.2 GeV) and (c) during unstable operation ($E_{\text{beam}} =$ 23.2 GeV) accompanied by heavy beam loss. From the figure we observe the following:

1) During injection (fig. 8(a)) two peaks show up. One is from 662 keV photons of the cesium source. The leftmost peak is the 30 keV K_α line of barium atoms in the cesium source which are excited by synchrotron radiation. Although this peak is prominent, 30 keV photons are absorbed in the first few mm of the crystal and will contribute little to the overall radiation damage. The continuum between both peaks is a superposition of inherent machine background radiation and photons from the cesium source which are backscattered into the BGO crystal.
2) During a quiet luminosity run (fig. 8 (b)) a similar picture emerges, however the Ba K_α line and the machine background with energies between 50 and 500 keV become more prominent.
3) The spectrum looks completely different during unstable beam conditions (fig. 8(c)). A broad background peak around 2 MeV is dominant. The intensity of this radiation is so strong that the yield from

* New measurements have shown: At a given length this dose reached all four sides of the crystal uniformly to within $\pm 15\%$

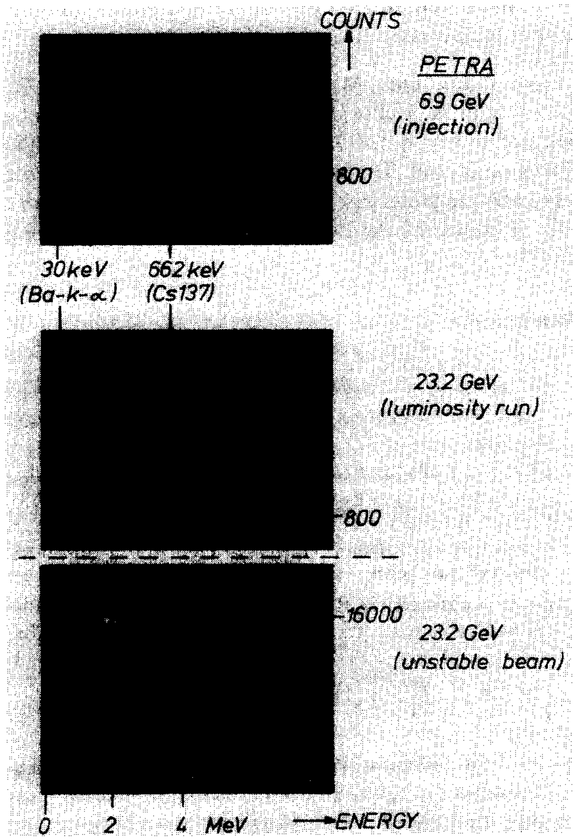

Fig. 8. Pulse height spectra measured by the BGO crystal under three different beam conditions. For details see the text.

662 keV photons is no longer visible.

The presence of a relatively energetic background radiation was confirmed by positioning several TLDs,

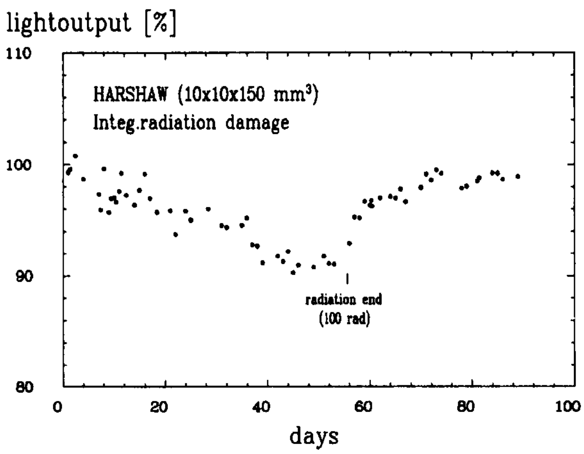

Fig. 9. Pulse height history of a 15 cm long BGO crystal exposed to background radiation on the PETRA beam pipe for 53 d.

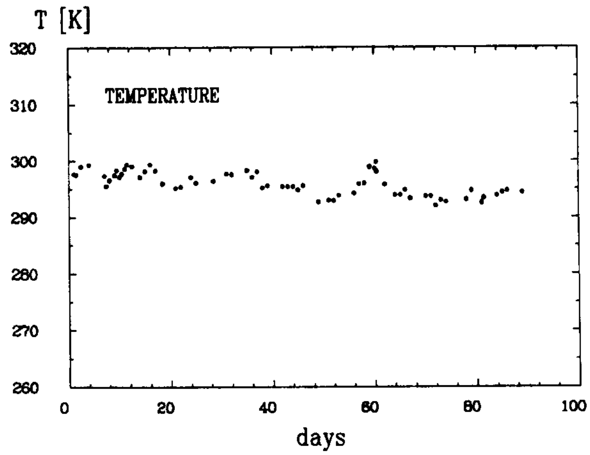

Fig. 10. Temperature history of the BGO crystal used in the test setup shown in figs. 6 and 7. Note that the results presented in fig. 9 have been corrected for electronic and temperature drift.

which were wrapped in lead layers of different thickness (between 1 and 10 mm) close to the BGO box. We conclude from the readings and from fig. 8 that the integrated radiation dose is primarily due to background radiation of photons and electrons with energies of a few MeV, which occur in times of very unstable high energy running and during beam dump after a luminosity run.

3.3. Results

Figs. 9 and 10 show the pulse height and temperature history of the crystal. The pulse heights shown are corrected for temperature and electronic drift. The measured points fluctuate by up to 2% depending on the smoothness of previous running. We observe a continuous decrease of the pulse height resulting in a final reduction of 9^{+1}_{-2} (syst.)% after 53 d **. After the end of the irradiation the crystal recovered with two time constants. It reached the original pulse height within 1% after about 20 d. The continuous pulse height decrease with a final reduction of 9% from daily doses of only 1.9 rad is unexpected. The damage is bigger than the conservative estimate of ref. [2], where an 1% asymptotic damage is expected from 0.9 rad d^{-1}.

4. Summary

We have tested the radiation resistance of BGO and CsI (Tl) crystals to low doses. From exposures of BGO crystals to 662 keV gamma radiation and to background

** We have confirmed this result in a second irradiation of 23 d, where the pulse height of the same crystal dropped by 7% at an integrated dosis of 75 rad.

radiation on the PETRA beampipe we observe remarkable pulse height reductions:

1) Exposures of the full crystal length of 15–20 cm to radiation of 85 and 40 rad (TLD readings) at rates of 1.7–4.7 rad/h^{-1} have lead to initial pulse height reductions of 25–38% and to permanent pulse height reductions of 1–13% with strong crystal to crystal variations.
2) Recovery times from a second exposure are shorter (full recovery within 10 d compared to 20–40 d after the first exposure). This indicates a partial annealing effect by repeated radiation.
3) Measurements on short crystals indicate that the long term damages observed in long crystals are mainly due to a reduction of the transmission properties.
4) A 53 d exposure of a 15 cm long BGO crystal on the PETRA beampipe at a rate of 1.9 rad/d^{-1} has led to a continuous decrease of pulse height with a final reduction of 9%. 33 d after the end of the exposure the crystal had recovered to the original pulse height within 1%. These variations indicate that the excellent resolution properties of BGO can only be exploited in a storage ring environment when continuous calibration to the 1% level is performed. We expect smaller damage in a detector (i.e. a barrel counter) where self-shielding protects the rear part of the crystal against low energy photons. Also reductions of transmission properties will be less harmful in crystals with wider cross sections than the ones used by us.

The exposure of a 10 cm long CsI(Tl) crystal to ^{137}Cs γ-ray doses of 42 and 85 rad has led to immediate pulse height reductions of 13 and 7%, respectively, suggesting that the damage saturates at a repeated exposure. No long term pulse height recovery is observed within errors of 2%, similar to experience with NaI(Tl) (see e.g. ref. [8]). Independent of the irradiation the crystal shows a 4%/month pulse height gain over a period of two months, probably due to a previous exposure to room light. Clearly the CsI crystal studied by us is not superior to typical BGO crystals concerning radiation resistance, pulse height stability and afterglow. Further work is required to relate the observed effects to crystal structure and impurities and to study the dependence of the damage on the transmission wavelength and the crystal temperature [18].

We thank Drs. M. Cavalli-Sforza, D. Coyne, E. Lorenz and R. Kellogg for suggestions and comments. We are indebted to Dr. E. Lorenz for supplying the Shanghai crystal. The help of Mr. K. Amsinck with the irradiation is gratefully acknowledged. Part of this work was supported by the Bundesministerium für Forschung und Technologie.

References

[1] L3 Collaboration, Technical Proposal (1983).

[2] F. Porter et al., UPSTATE: A Proposal for a High-Precision North Area Detector, submitted to Cornell University (1983).

[3] CELLO Collaboration, Proposal to Upgrade the CELLO Detector, Internal Report, DESY-CELLO-83-01 (1983).

[4] ARGUS Collaboration, A Proposal to study $\gamma\gamma$ Interactions with the detector ARGUS at DORIS, Report DESY PRC.83/06 (1983).

[5] A.C. Lucas and M.R. Mayhugh, Characteristics of Bismuth-Germanate crystals after Extended Irradiation, Harshaw Report (1977) unpublished; K. Takagi et al., J. Crystal Growth 52 (1981) 584.

[6] M. Kobayashi et al., Nucl. Instr. and Meth. 206 (1983) 107.

[7] M. Cavalli-Sforza, Proc. Int. Workshop on Bismuth-Germanate, ed., C. Newman-Holmes, Physics Department Report, Princeton University (1982).

[8] G.J. Bobbink, A. Engler, R.W. Kraemer, J. Nash and R.B. Sutton, Carnegie Mellon University Report CMU-HEP 83-13 (1983).

[9] Harshaw Chemie BV, NL 3454 PK De Meern, Holland.

[10] Crismatec, F 38610 Gieres, France.

[11] Nihon Kessho Koogaku Co, 318-19 Kondo, Tatebayashi City, Japan.

[12] Shanghai Institute of Ceramics, Ning Nang Road 862, Shanghai, China.

[13] IBS, D-8082 Grafrath (produced in England).

[14] Dr. Korth oHG, Am Jägersberg 9, D-2300 Kiel (produced by BDH, England).

[15] H. Genzel, RWTH Aachen, private communication.

[16] M.R. Farukhi, IEEE Trans. Nucl. Sci. NS-29 (1982) 1237.

[17] K. Wittenburg, diploma thesis, Universität Hamburg (1984) unpublished.

[18] For recent measurements see: C. Laviron and P. Lecoq, CERN/EF 84-5 (1984).