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Radiation resistance and optical properties of lead fluoride Cherenkov crystals

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Abstract

Optical properties of large size lead fluoride (PbF_2) crystals of three different manufacturers and their degradation caused by ^{60}Co γ-radiation have been investigated. Transmission losses have been systematically studied at absorbed energy doses between 0.1 and 7 kGy. Several radiation induced absorption bands have been observed. Optical bleaching with light of wavelengths $\gtrsim 365$ nm has been found very effective to restore the original characteristics even after repeated irradiations. This observation together with the high density and the ultraviolet extended transmission make PbF_2 an excellent choice for high rate and high resolution e.m. calorimetry.

1. Introduction

Lead fluoride (PbF_2) crystals have first been studied as a possible material for electromagnetic calorimetry in 1968 by Dally and Hofstadter [1]. In 1990 Anderson [2] confirmed the absence of any scintillation component in the PbF_2 light output, identifying it as a pure Cherenkov radiator. This material with a density ρ of 7.77 g/cm^3, a radiation length X_0 of 0.93 cm and a Molière radius R_M of 2.2 cm (apparent $R_M \approx 1.8$ cm [2]) is suitable for large scale industrial production. Because of its compactness and superior timing properties it has drawn increasing attention in the last few years, especially since it had been considered for the SSC experiment GEM. The parity violation experiment A4 [3], which is currently being set up at the Mainz microtron (MAMI), will comprise 1022 PbF_2 crystals. The smallness ($\approx 10^{-6}$) of the parity violation asymmetry requires a very high luminosity ($> 0.5 \times 10^{38}$ cm^{-2}s^{-1}) and the capability to detect a 10 MHz signal rate of elastically scattered electrons in the presence of a 100 MHz background of photons, soft electrons, pions and protons. To control pile-up losses carefully during the measurement of ppm asymmetries, a PbF_2 calorimeter offers the best perspective.

The total absorbed dose in a single crystal of the 0.7 sr solid angle calorimeter within a 1000 hours measurement period has been estimated to be of the

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order of 100 Gy [4]. While PbF_2 fulfills stringent demands on energy resolution ($\Delta E/E = 3.2\%/\sqrt{E[\mathrm{GeV}]}$) and timing (time response < 20 ns) [5], its radiation resistance remained an unresolved key question. A previous study of the radiation resistance of PbF_2 crystals from Optovac [6] has been made by Woody [7]. It was indicated that those crystals were about 10 times less sensitive to radiation than lead glass and that most of the damage is reversible by bleaching with blue light. Further study of the radiation resistance of PbF_2 crystals is not only relevant for the A4 experiment but it is of general interest, since very high absorbed doses are expected for experiments at the new generation of high luminosity colliders. As an example the expected radiation dose in the end-cap of the CMS calorimeter (at LHC) is about 70 kGy for an integrated luminosity of $5\times10^5\ \mathrm{pb}^{-1}$, corresponding to the first ten years of operation [8]. Provided that the radiation damage in PbF_2 can be controlled, this material is very interesting for high event rate calorimeter applications.

Radiation damage in glasses and single crystals has been studied extensively [9]. Concerning the optical properties of these materials only the transmittance is affected in the case of a pure Cherenkov radiator. Fig. 1 provides an overview of the degradation of the transmittance induced by 200 Gy of γ-irradiation in the following samples:

1. a $40^2\times290\ \mathrm{mm}^3$ block of lead glass (SF5),
2. a $40^2\times130\ \mathrm{mm}^3$ lucite Cherenkov radiator and
3. a $30^2\times160\ \mathrm{mm}^3$ PbF_2 crystal grown by SICCAS [10].

Transmittance curves before and after irradiation are presented. Details of the irradiation procedure, the measurement technique and the corrections applied will be given in Sections 2 and 3. The transmission losses can be characterized by the change of the transmittance $\tau^{\mathrm{i}}(\lambda)$ through one radiation length or by the change of the absorption length $\Lambda_{\mathrm{Abs}}(\lambda)$. In PbF_2 a decrease of Λ_{Abs} from 41.8 to 10.9 cm has been found at 380 nm which is about one order of magnitude less than in lead glass (decrease from 16.1 to 3.8 cm). Fig. 1 shows that only PbF_2 transmits in the ultraviolet region below 350 nm. On the one hand this is an advantage due to the quadratic increase of the Cherenkov light yield with decreasing wavelengths. On the other hand it is in this part of the spectrum where considerable radiation damages have been found in PbF_2.

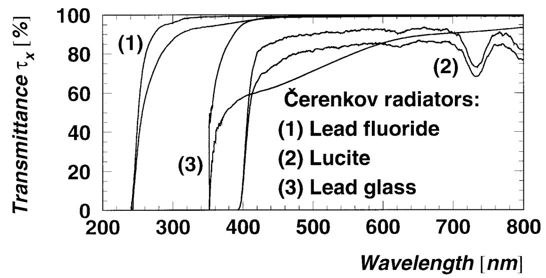

Fig. 1. Transmission and radiation resistance of PbF_2 in comparison to other Cherenkov radiators. The internal transmittance $\tau^{\mathrm{i}}_{\mathrm{X}}$ before and after absorption of a 200 Gy dose from ^{60}Co-radiation is shown.

2. Optical properties before irradiation

The transmission of various samples has been investigated by measuring the optical transmittance factor τ, which is the transmitted light flux Φ_{T} normalized to the incoming flux Φ_0. For this a commercial double beam spectrophotometer Shimadzu UV-2101 PC was used. The measured transmittance τ has to be corrected for reflection losses in order to obtain the internal transmittance $\tau^{\mathrm{i}}(\lambda) = (1/P)\tau(\lambda)$. The reflection factor $P = (4n)^2/(1+n)^4$ can be determined by the Fresnel equation $R = ((n-n')/(n+n'))^2$ assuming normal incident light and two parallel end surfaces, where n and n' are the refractive indices of air and PbF_2 ($n \approx 1.7$–1.9).

Several PbF_2 crystals from the following three different manufacturers have been investigated: Korth [11], Optovac [6] and SICCAS [10]. In Fig. 2 the internal transmittance $\tau^{\mathrm{i}}_{\mathrm{X}}$ for one radiation length measured through the longitudinal axis is presented. This figure shows considerable differences between the transmission of crystals from the different manufacturers. All SICCAS crystals exhibited a weak absorption band at 300 nm;

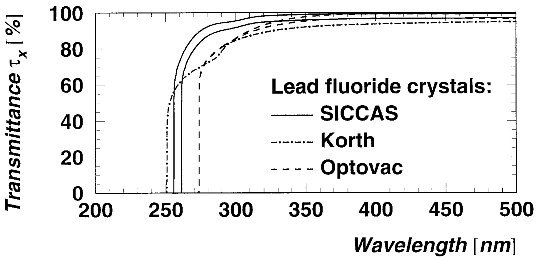

Fig. 2. A comparison of the internal transmittance τ^{i}_{X} through the longitudinal axis for PbF_2 crystals from three different manufacturers. For SICCAS and Optovac, crystals with the highest and lowest transparency are shown.

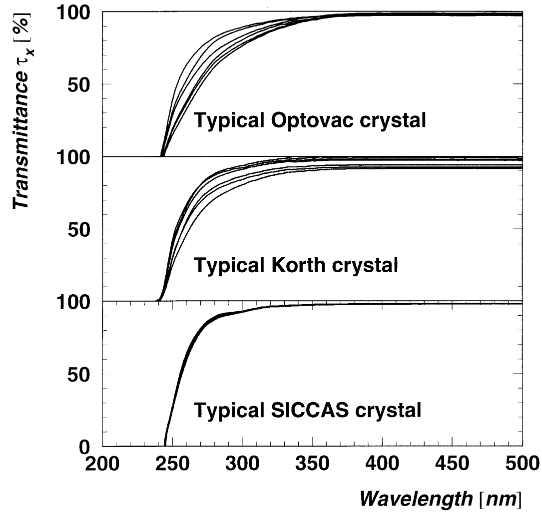

Fig. 3. Homogeneity of PbF_2 crystals. The internal transmittance τ^{i}_{X} perpendicular to the longitudinal axis at six positions is shown for crystals from three different manufacturers.

in the Korth crystals a band occurred at ≈ 285 nm; no bands were visible in the Optovac crystals. For crystals from the same manufacturer only small differences have been observed. In most cases the poor transmission of some crystals appears not to be connected with macroscopic imperfections in the bulk material. The transmission of the best crystals from both Optovac and SICCAS in the visible region was very close to the theoretical limit for perfectly polished surfaces without absorption. Table 1 summarizes crystal sizes and internal transmittancies τ^{i}_{X} at 285 and 300 nm as well as the corresponding light absorption lengths.

The determination of τ^{i} through the longitudinal axis of the crystal is not sufficient to characterize them completely. For detector applications the spacial homogeneity of the transmission is of crucial importance, too. Significant variations of the transmittance were found in crystals from two manufacturers (Korth and Optovac) while crystals from SICCAS exhibited virtually no variations. The transmission perpendicular to the longitudinal axis measured at six different positions is shown in Fig. 3. At a typical wavelength of 300 nm the transmittance was found to depend linearly on the position with a spatial gradient $\alpha = \Delta\tau^{i}_{X}/\Delta z = 2.4\%/\mathrm{cm}$ in the Korth crystal and $\alpha = 1.5\%/\mathrm{cm}$ in the Optovac crystal, respectively. At wavelengths $\gtrsim 350$ nm only the crystal from one manufacturer showed variations along the axis. This may reflect the different growth methods of the crystals. The SICCAS

Table 1
Properties of investigated PbF_2 crystals. The internal transmittance τ^{i}_{X} and the light absorption length Λ_{Abs} are given for the two SICCAS and Optovac crystals with the highest and lowest transparency, respectively, and for one of the Korth crystals

Manufacturer	Size (mm^3)	Crystal	τ_x (285 nm) (%)	Λ_{Abs} (285 nm) (cm)	τ_x (300 nm) (%)	Λ_{Abs} (300 nm) (cm)
SICCAS	$30^2 \times 160$	Worst	89.4	8.9	93.9	15.9
		Best	92.1	12.1	95.6	22.2
Optovac	$21^2 \times 185$	Worst	78.8	4.2	86.1	6.7
		Best	78.7	4.2	86.3	6.8
Korth	$15^2 \times 100$		75.1	3.5	82.5	5.2

crystals have been grown by a modified Bridgeman technique [12] in contrast to the crystals from Korth and Optovac, but unfortunately we are not given the orientation of the seed side in the ingot of the samples.

3. Optical properties after irradiation

We have investigated the radiation resistance of PbF_2 to γ-radiation of a ^{60}Co source at various absorbed energy doses. Irradiations have been performed at the 55 TBq facility of the Strahlenzentrum at the Justus–Liebig-Universität Gießen, Germany. The experimental set-up is schematically shown in Fig. 4. Depending on the chosen distance between source and sample, a power between 50 and 300 mGy/s was absorbed, causing no observable thermal effects. The samples were fixed with their longitudinal axis perpendicular to the direction of the γ-quanta and to the symmetry axis of the source. Typical irradiation times varied between 0.5 and 5 h corresponding to an energy dose of 0.1–7 kGy.

Due to the finite size of the samples a depth dependence of the absorbed energy was expected. Therefore the variation in the irradiation effect has been investigated by measuring the transmittance in Y-direction at different X-positions. A variation of less than 4% along the width of the samples has been observed, which can be explained by the dominance of the Compton effect in this energy range.

For some SICCAS crystals radiation damages in the transmittance perpendicular to the longitudinal axis at two different longitudinal positions are displayed in Fig. 5. A significant increase of the transmission losses with the absorbed energy dose is clearly visible. In Fig. 6 results for two Korth crystals are presented. Measurements for several longitudinal positions, indicated by different line types, are shown, since considerable variations have been observed both before and after irradiation. The striking conclusion from the curves is that an initial variation in the transmittance is further enhanced by radiation. An alternative representation of the radiation damage can be given by the ration R, which is the transmittance τ_X^i after irradiation normalized to the initial transmittance. For the Korth crystals this ratio is strongly dependent on the position and varies between the two front faces of the crystals by as much as 0.16–0.21 at 300 nm for a dose of 7 kGy. In contrast, the SICCAS crystals exhibited no spatial dependence of this ratio.

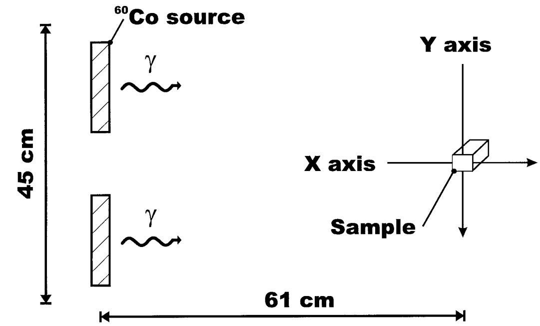

Fig. 4. Sketch of the experimental set-up for irradiating various samples. The X- and Y-direction of the transmittance measurements are defined in respect to the photon flux of the ^{60}Co source.

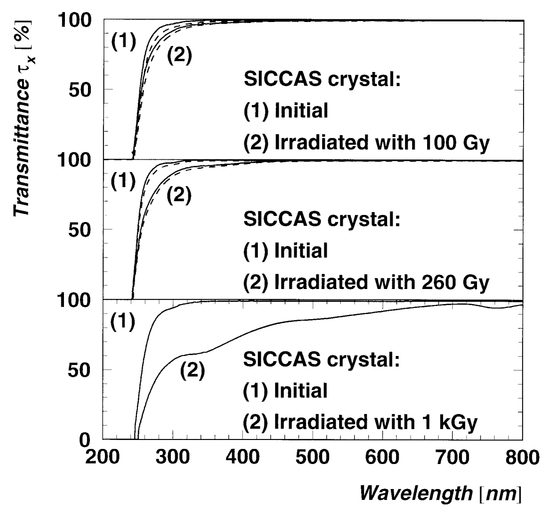

Fig. 5. Spatial dependence of the radiation damage in different PbF_2 crystals from SICCAS. The internal transmittance τ_X^i perpendicular to the longitudinal axis before and after γ-irradiation with different doses is shown. Top: 1 kGy. Bottom: 7 kGy. The longitudinal positions are encoded as follows: solid = 1 cm; dashed = 5 cm.

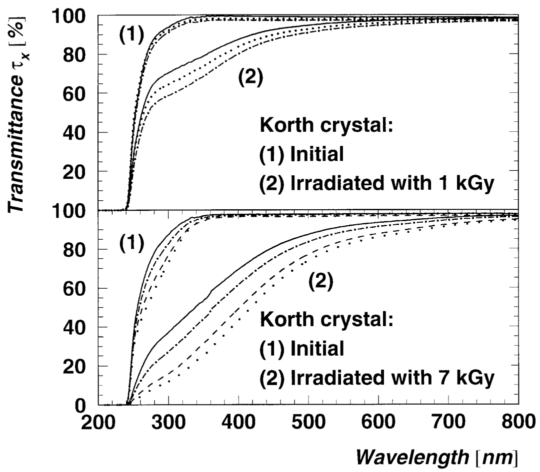

Fig. 6. Spatial dependence of the radiation damage in different PbF_2 crystals from Korth. The internal transmittance τ^i_X perpendicular to the longitudinal axis before and after γ-irradiation with different doses is shown. Top: 1 kGy. Bottom: 7 kGy. The longitudinal positions are encoded as follows: solid = 1 cm; narrow dots = 2 cm; dot-dashed = 3 cm; dashed = 6 cm; wide dots = 9 cm.

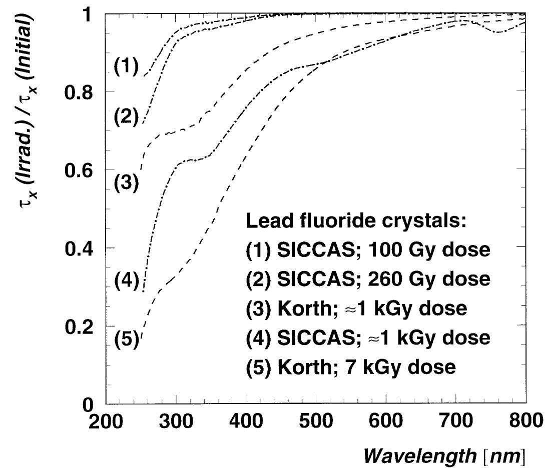

Fig. 7. Damage in PbF_2 crystals due to γ-irradiation. The ratio R given by the internal transmittance τ^i_X after irradiation with various doses normalized to the initial transmittance is plotted. The curves (1, 2, 4) represent SICCAS crystals whereas the curves (3, 5) represent values for a central position in Korth crystals.

In Fig. 7 the ratio R is shown for different manufacturers and energy doses. For the Korth crystals we restricted ourselves in this figure to a central position. In addition to the general trend of an increase of the damages with the absorbed dose, concentrated in the UV region below 300 nm, some special features can be noted. Due to the different radiation conditions, crystal geometries and the observed spatial variation in the sensitivity of the Korth crystal the differences between curves (3) and (4) are not considered to be indicative of a difference in the radiation resistance of the two crystals. Furthermore a common absorption band seems to be present at 350 nm, while only the SICCAS crystals exhibit broad absorption structures around 500 and 765 nm. Shoulders in the deep ultraviolet transmission which indicate additional, strong absorption bands occured at ≈ 300 nm in the SICCAS crystals while they are observed at ≈ 270 nm in the Korth crystals.

It should be stressed that even the apparently mild degradation of the optical properties after absorbed doses of 100 Gy would result in a significant loss of photons and a deterioration of the energy resolution of PbF_2 detectors.

4. Optical bleaching of crystals

After irradiation, a spontaneous recovery of the transmittance at room temperature has been observed. In a SICCAS crystal, irradiated with a dose of 100 Gy, an increase from 91% to 93% at 300 nm has been found after 15 days of storage. For the heavily irradiated Korth crystals a recovery of 10–15% was reached after only 4 days. Some further restoration of the transmittance in the entire spectrum was seen on a time scale of a few weeks.

This process can be enhanced by illuminating the damaged crystals with blue light. For this optical bleaching we have employed a low-pressure mercury(argon) pencil lamp. Its irradiance was calculated to be $\approx 2\,\mu W/cm^2$ at a distance of 50 cm. Successful bleachings have been performed with this mercury lamp filtered at 365 nm and with quartz tungsten halogen (QTH) lamps. The crystals have been illuminated and their transmittance has been measured frequently until a saturation value was reached. In all crystals substantial restoration of transmission was achieved, nevertheless a small residual damage remained even after prolonged illuminations. As it can be inferred from Fig. 8 the

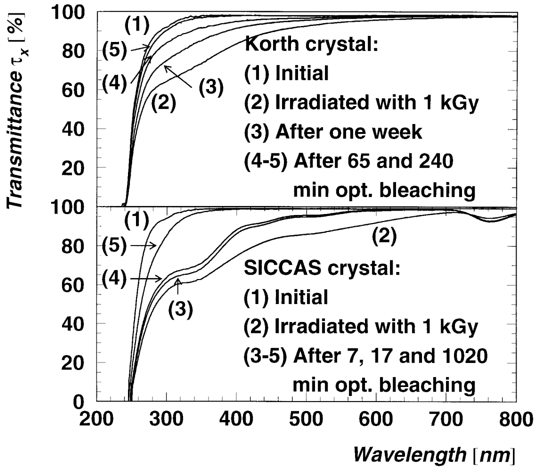

Fig. 8. Reduction of the initial (1) internal transmittance τ^i_X induced by γ-irradiation (2) and its restoration (3–5) in PbF_2 crystals. Optical bleaching was performed with two different lamp sources. Top: a filtered mercury lamp. Bottom: a quartz tungsten halogen lamp.

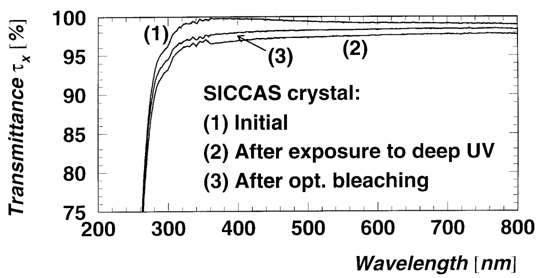

Fig. 9. Decrease of the apparent internal transmittance τ^i_X in PbF_2 caused by 40 min of illumination with deep ultraviolet light. Curve (3) shows the effect of 20 h of optical bleaching with blue light.

time dependence of the recovery in different parts of the spectrum is complex. Only in regions without strong absorption bands it appears to be purely logarithmic. It is interesting to note that repeated irradiations after optical bleachings exhibited no increased sensitivity for absorbed doses up to 100 Gy.

In order to reduce the residual damage the mercury lamp was utilized without the filter, i.e. the harder UV light of the intense 253.7 nm line was added, but no improvements have been found. Furthermore, a new, previously not irradiated crystal, was exposed to the light. In Fig. 9 it is shown that the internal transmittance τ^i_X between 250 and 800 nm decreased by 1–3% after 135 min of illumination. The obvious explanation to this investigation is the strong lattice absorption in the UV of the small layer near the surface of the material. Optical bleaching with light > 365 nm for 20 h only slightly improved the transmittance. This lead to the conclusion that deep ultraviolet light damages the crystal surface permanently.

5. Summary

The optical quality of PbF_2 crystals of different manufacturers has been investigated. Crystals grown by SICCAS showed a $\approx 10\%$ larger internal transmittance τ^i_X at wavelengths < 300 nm than those grown by Optovac and Korth. In addition, the transversal transmittance of SICCAS crystals was almost homogeneous along the longitudinal axis, whereas the other crystals exhibited spatial variations. They may be attributed to the growth conditions of the crystals, particularly to the segregation of impurities. A spatial gradient in the transmission of detector crystals would reduce the energy resolution of PbF_2 calorimeters.

It was confirmed that lead fluoride crystals are about ten times more radiation resistant than lead glass detectors. Significant transmission losses have been observed, however, after irradiation with ^{60}Co photons. Some absorption bands appeared in the transmission curves. It was found that crystals from different manufacturers showed considerable differences in their sensitivity to radiation. An enhancement of the spatial gradients in the transmittance has been observed in Korth crystals.

In damaged crystals some small spontaneous recovery at room temperature was observed. An almost complete recovery could be induced by illuminating the crystals with blue light. Recovery even after excessive doses of 7 kGy could be achieved within a few hours of bleaching utilizing commercial mercury or QTH lamps.

In conclusion PbF_2 is extraordinarily well suited for e.m. calorimetry even in severe radiative environments provided that regular bleaching processes are employed. For the forthcoming A4 calorimeter blue light will be accommodated via fibre guides by QTH lamps.

Acknowledgements

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