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A visualization of the damage in Lead Tungstate calorimeter crystals after exposure to high-energy hadrons

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ABSTRACT

The anticipated performance of calorimeter crystals in the environment expected after the planned High-Luminosity upgrade of the Large Hadron Collider (HL-LHC) at CERN has to be well understood, before informed decisions can be made on the need for detector upgrades. Throughout the years of running at the HL-LHC, the detectors will be exposed to considerable fluences of fast hadrons that have been shown to cause cumulative transparency losses in Lead Tungstate scintillating crystals. In this study, we present direct evidence of the main underlying damage mechanism. Results are shown from a test that yields a direct insight into the nature of the hadron-specific damage in Lead Tungstate calorimeter crystals exposed to 24 GeV/c protons.

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1. Introduction

Detectors now in operation at the Large Hadron Collider at CERN may need to face an increasingly challenging environment after the accelerator upgrade to High-Luminosity running (HL-LHC), that is being planned to start – according to the present schedule – in 2022. While performance requirements will be driven by the physics results obtained at the LHC in the near future, a thorough understanding of the detector behavior is needed, as an input to knowledgeable decisions concerning upgrades.

In this framework, our earlier studies on Lead Tungstate (PbWO₄) [1–3] have established that hadronic showers from high-energy protons [1] and pions [2] cause a cumulative loss of Light Transmission in PbWO₄, which is permanent at room temperature, while no hadron-specific change in scintillation emission [3] was seen. In Ref. [2] we have also measured and compared the damage amplitudes for 24 GeV/c protons and for 290 MeV/c charged pions, and we have shown how they can be rescaled to different running conditions.

The cumulative nature of the damage implies that the light output from Lead Tungstate crystals used in a calorimeter will decrease during operations in proportion to their exposure to an integrated flux of energetic hadrons. The acceptable performance, thus tolerable amount of damage, depends on the physics goals of the experiment, in that the change of light output is expected to affect the achievable energy resolution. The groundbreaking results published in Refs. [1–3] have been used in Ref. [4] e.g. to extract expectations for the loss of light output in the CMS electromagnetic calorimeter as a function of the integrated luminosity delivered by the Large Hadron Collider. Further, the CMS Collaboration has started a series of tests aimed at quantifying the achievable energy resolution for electrons and photons as a function of hadron fluence [5,6]. These effects are also addressed in the CMS Upgrade Technical Proposal [7] that mentions how the Collaboration has started an extensive program of simulations and R&D to assess the expected performance evolution and to identify suitable materials, were a calorimeter upgrade for the HL-LHC running to be needed. A thorough understanding of the damage mechanisms at play is clearly vital and has motivated the work presented herein.

The features observed in hadron-irradiated Lead Tungstate, in fact, hint at local centers of damage that might be caused by fragments of the heavy elements, Pb and W as the dominant cause of transmission losses. Such fragments can have a range up

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to 10 μm and energies up to ~ 100 MeV, corresponding to a stopping power four orders of magnitude higher than the one of minimum-ionizing particles [1]. The qualitative understanding we gained of hadron damage in Lead Tungstate led us to predict [8] that such hadron-specific damage contributions are absent in crystals consisting only of elements with $Z < 71$, which is the experimentally observed threshold for fission [9], while they should be expected in crystals containing elements with $Z > 71$. We confirmed the first prediction with measurements [10] that show how hadrons in Cerium Fluoride cause a damage that recovers at room temperature, with none of the features we observed for Lead Tungstate. The second prediction is confirmed by existing proton-damage measurements in BGO [11,12], in Lead Fluoride and BSO [13], which all contain elements with $Z > 71$.

All this indirect evidence is consistent with a mechanism by which the heavy fission fragments deposit a lot of energy along their short track, leaving regions within the crystal where the lattice structure is modified: it can remain disturbed, strained, disordered, or re-oriented. These damage regions have different optical and mechanical properties from the surrounding crystal lattice, and thus they can act as scatterers for light propagating in the crystal, whatever its origin, from scintillation or from an external source.

The mechanism is well known since a long time, and is commonly called “fission track damage” in the literature. In fact, almost a century ago, Dessauer formulated the concept of a “thermal spike” when an incident ion comes to a stop in matter [14]. Later on, Chadderton and Torrens, in their book on the subject [15], explained how “Along the heated cylindrical track of the fragment the crystalline matter is disturbed, decomposed, or removed. The subsequent arrangement is not necessarily perfect and strain centers or dislocations remain” and ion implantation has become, in recent days, a technique used to change material properties [16].

In this study we present direct, visual evidence that confirms our understanding of hadron damage mechanisms. This will help making informed decisions regarding future calorimeter designs or upgrades, to ensure an optimum long-term detector performance.

2. Macroscopic observations

The features of hadron-induced changes in Light Transmission published in Ref. [1] are the peculiar ones of Rayleigh scattering, which implies the presence of dipole-shaped regions acting as scatterers for the light. Here, we achieve the visualization of such scattering effects by shining green 543.5 nm LASER light through a hadron-irradiated crystal, as exemplified in the photograph of Fig. 1. The light gets scattered, making the beam visible. In the same photograph, a Polaroid filter was introduced between the observer and the crystal, as indicated by the dashed rectangle, so as to cover the right half of the crystal: the scattered LASER light is not transmitted by the filter. This demonstrates how the

scattered light is polarized, yet another feature that is distinctive for Rayleigh scattering, and thus for the presence of localized regions of damage acting as scatterers for the light. All of these features are also observed at different wavelengths, as verified with 633 nm LASER light.

3. Microscopy observations—the method

While the macroscopic observations in Section 2 reveal the presence of scatterers, these are not individually visible to the naked eye. It was thus interesting to pursue a microscopy visualization, following the fission track analysis method used in geochronology for mineral dating [17]. The method is commonly used on minerals containing Uranium, which naturally exhibit damage tracks caused by the daughter products from spontaneous fission of ^{238}U . Geochronological mineral dating relies on the count of these fission tracks, but it also needs a complementary information: the Uranium concentration at a given time. Based on the $^{235}\text{U}/^{238}\text{U}$ abundance ratio, the Uranium concentration can be determined by inducing ^{235}U fission in the studied mineral through thermal neutron irradiation, with an external detector, such as high-purity muscovite mica, held in intimate contact with a polished section of the mineral surface. The ^{235}U fission fragments create induced tracks that reach the overlying external detector, where they are later revealed by chemical etching to make them easily visible for counting under an optical microscope and thus infer the ^{238}U concentration. It was thus obvious to apply this well-established methodology to attempt revealing induced fission damage in Lead Tungstate.

4. The irradiation setup

We have performed hadron irradiations using a setup suitable for allowing the visualization of damage trails, using the 24 GeV/c proton beam at the IRRAD1 facility [18] located in the T7 beam line of the CERN PS accelerator.

For each irradiation, two samples have been prepared, where a Lead Tungstate slide, 2 mm thick and $1\text{ cm} \times 2\text{ cm}$ in cross-section, has been placed in intimate contact with a high-purity muscovite mica ($\text{KAl}_2(\text{AlSi}_3\text{O}_{10})(\text{F},\text{OH})_2$) slide of same cross-section, 0.1 mm thick. The contact was ensured by a heat-shrinkable plastic wrapping. The samples have been placed behind a 7.5 cm long Lead Tungstate crystal for the irradiation, to make sure the running conditions are reproduced, that are encountered in homogeneous high-energy physics calorimetry, where energetic hadrons cause penetrating hadron showers, i.e. a cascade of nuclear interactions which are at the origin of the observed hadron-specific damage. The schematics of the irradiation setup is visible in Fig. 2. For the hadron irradiation, the proton beam was broadened to cover the whole crystal cross-section, and the fluence for each irradiation was determined

Fig. 1. Laser light (here at 543.5 nm) is scattered, and thus becomes visible, when shone through a hadron-irradiated Lead Tungstate crystal. A polaroid filter (indicated by the dashed line) blocks the scattered light, revealing its linear polarization.

following the method described in Ref. [1], from the activation of a high-purity aluminum foil covering the crystal front face.

Three irradiations were performed, on different pairs of samples, up to fluences of, respectively, $\Phi_p^1 = (1.17 \pm 0.09) \times 10^{11} \text{ cm}^{-2}$, $\Phi_p^2 = (1.59 \pm 0.13) \times 10^{12} \text{ cm}^{-2}$ and $\Phi_p^3 = (0.73 \pm 0.09) \times 10^{13} \text{ cm}^{-2}$.

5. Visualization of damage tracks in mica

Fission damage in mica changes its crystalline structure into a metamict state, that is easily etched according to a well-known recipe [19], whereby the slides are immersed in HF at 40% for 45 min at 20 °C. The procedure allows to remove the material of the damaged region, leaving the surrounding crystalline matter intact. The possible track topologies in an etched mineral are illustrated in Fig. 3(a). Examining by eye the mica slides after chemical etching (Fig. 4), one notices a coloration that intuitively correlates with the applied irradiation fluence. The mica slides were then examined with an Axio Imager Z1M optical microscope from Zeiss [20]. Transmitted light images of the surface that was in intimate contact with the Lead Tungstate during irradiation, are shown in Fig. 5. Linear damage tracks are visible, and they were counted at 1000 $\times$ magnification except for the highest fluence, where individual tracks could not be visually resolved with accuracy. In contrast, no tracks were observed on the opposite surface, which was not in contact with the Lead Tungstate. This by itself, is sufficient evidence that all tracks come from the Lead Tungstate, and none of them originate from inside the mica.

The track densities in Fig. 5 clearly correlate with the irradiation fluences, as listed in Table 1. Even where, for the highest fluence, no accurate count was possible, the proportionality is visually striking.

Fig. 2. Irradiation setup for fission-track visualization (see Section 4 for a detailed description).

It can be qualitatively observed that the damage tracks are straight and occur in random orientations, as expected due to fission fragments produced by interactions in a hadron shower, and that they do not follow any pre-existing fabric in the mica.

Additional evidence for the track origin is supplied by images obtained in reflected light (Fig. 6). In such images, the entrance point of each track is visible as a dark spot (“etch pit”), while in transmitted light images it is obscured by the track shadow. The presence of an etch pit at the extremity of all tracks implies that their origin is external to the mica. In fact, the only topology that could allow for the observation of a track without an associated etch pit is the intersection between a track coming from the surface with one fully contained in the mica. The latter would thus be etched as well and become visible, as illustrated in Fig. 3(a). The observed topologies in Figs. 5 and 6 all correspond to tracks associated with an etch pit, as illustrated by Fig. 3(b). No evidence of confined tracks is found.

From all these observations, the origin of the observed tracks is clearly established to lie in the projection of break-up fragments from nuclei in the Lead Tungstate that underwent fission, according to the mechanisms explained in Section 1.

6. Complementary tests on Lead Tungstate

The methods developed in geochronology, to use mica as an external detector, have allowed us to avoid having to etch the activated Lead Tungstate slides. An observation of those slides was however attempted, without a surface treatment, in a Sigma scanning electron microscope from Zeiss [20] equipped with a HKL Advance electron backscattered diffraction system from Oxford Instruments [21]. For each surface element hit by the

Fig. 4. Mica slides, after chemical etching, that were irradiated up to three different fluences in intimate contact with Lead Tungstate, as an external detector for fission tracks.

Fig. 3. Illustration (a) of all possible track topologies in an etched mineral and (b) of the features observed in the present study.

Fig. 5. Transmitted-light optical-microscopy images of fission tracks in the mica slides from Fig. 4 for the three irradiation fluences, Φ_p^1 (top), Φ_p^2 (middle) and Φ_p^3 (bottom).

electron beam during the scanning, an electron diffraction pattern is obtained and the local crystallographic orientation calculated from it. The quality of the pattern is influenced by a number of factors including local crystalline perfection, to account for it a quality value according to the contrast and sharpness of the diffraction pattern is also attributed to each surface element. Orientation maps and quality value maps provide a useful

Table 1
Densities of observed tracks counted on the mica slides. No accurate count was possible at the highest fluence.

p fluence (10^{12} cm^{-2})	Track density (cm^{-2})	Ratio (10^5)
0.12 ± 0.01	$(2.9 \pm 0.1) \times 10^5$	4.0 ± 0.5
1.59 ± 0.13	$(2.8 \pm 0.1) \times 10^6$	5.6 ± 0.5
7.3 ± 0.9	Not counted	–

visualization of regions with different lattice orientation and with disturbed lattice structure, respectively, but is limited by the lateral resolution of the technique estimated to be of 200 nm (size of the smallest distinguishable details obtained in a scratched sample of the same material using the same analysis conditions). In order to avoid any artifact resulting from lapping and polishing micro scratches, cleaved faces obtained by breaking irradiated and not-irradiated slides were examined. As expected from a single crystal free of lattice damage, the orientation and quality value in the non-irradiated sample were homogeneous throughout the mapped region. The same results were obtained from the irradiated sample. This is not a surprise: typical fission track dimensions are small compared with the resolution of the technique.

All of these features are compatible with the observation of Rayleigh scattering in Lead Tungstate and of tracks due to fission fragments in mica, due to a mechanism where

- (a) the fission fragments damage the crystal through their extremely high energy loss over a short path;
- (b) damage is followed by an immediate recrystallization [22];
- (c) optical boundaries, which act as scatterers, are due to mechanical strains remaining after re-crystallization [23].

It was in fact noticed e.g., by Mills that Rayleigh-type light scattering can occur simply due to the damage regions having “optical boundaries” from mechanical strains after re-crystallization: “...the inclusion of scattering from the strain halo ...leaves the well-known ω^4 dependence unaltered” [24].

It might be interesting to note that heating the crystals up to 350 °C after ingot production allows to anneal mechanical stresses, which is a standard procedure industrially used before cutting and polishing [25]: thus, we can establish a link between light scattering and mechanical strains by applying to a proton-irradiated crystal the same heat treatment just described.

The progressive recovery of Light Transmission for a crystal previously irradiated with protons through heating up to different temperatures is visible in Fig. 7, where transmission curves are shown after the crystal was subjected to various heating cycles performed in an increasing temperature order. We annealed the crystal, labeled “d” in Ref. [1], 240 days after irradiation. The annealing temperature was reached in a programmable oven, by increasing the temperature linearly over 2 h. The crystal was then kept at the target annealing temperature for 8 h. The temperature of the oven was then reduced to room temperature over two hours. While essentially no recovery of the crystal is observed at 160 °C, partial recovery takes place at 250 °C. At a temperature of 350 °C almost complete recovery is reached, except for a small residual shift of the band-edge. Shining LASER light through the crystal after the damage annealing by heating, no scattering centers are observed. The outcome of this test qualitatively supports the understanding that the scattering centers present in proton-irradiated Lead Tungstate crystals are due to mechanical strains left by the recrystallization following hadron damage, and are thus eliminated by the same heat treatment that is effective in annealing such strains.

Fig. 6. Reflected-light optical-microscopy images of fission tracks in the same mica slides regions as in Fig. 4 for the three irradiation fluences, Φ_p^1 (top), Φ_p^2 (middle) and Φ_p^3 (bottom).

7. Conclusions

Macroscopic observations show that light scattering centers are left in Lead Tungstate after hadron irradiation, which are permanent at room temperature, as described in our earlier work [1]. The work presented here has allowed to visualize this damage at a microscopic scale, as damage tracks originating in

Fig. 7. Transmission curves for a crystal, 23 cm long, that was irradiated with protons up to a fluence $\Phi_p = (1.32 \pm 0.11) \times 10^{13} \text{ cm}^{-2}$, after heating in air up to different temperatures.

Lead Tungstate and entering mica slides used as an external detector. Scanning Electron Microscope imaging tests indicate the damage regions in Lead Tungstate are in average crystalline but for – possibly – a track core, that is not visible due to the finite spatial resolution of the technique. It is understood that Rayleigh-type light scattering can occur, due to the damaged regions having “optical boundaries” from mechanical strains remaining after re-crystallization.

The microscopic observations presented in this work confirm our understanding of the damage mechanism due to hadrons in Lead Tungstate, where the fragments from the fission induced in Lead and Tungsten cause severe, local damage to the crystalline lattice due to their extremely high energy loss over a short track, a behavior which is common to scintillators composed by elements with a $Z > 71$, which is the threshold [9] at which fission becomes important. This evidence is relevant, in that it confirms our understanding of the damage mechanisms involved.

The effects that have been visualized with this work need to be taken into account when selecting materials for high-energy physics calorimeters, where an exposure to large integrated fluences of energetic hadrons is expected.

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References

- [1] M. Huhtinen, P. Lecomte, D. Luckey, F. Nessi-Tedaldi, F. Pauss, Nuclear Instruments and Methods in Physics Research Section A 545 (2005) 63.
- [2] P. Lecomte, D. Luckey, F. Nessi-Tedaldi, F. Pauss, D. Renker, Nuclear Instruments and Methods in Physics Research Section A 587 (2008) 266.
- [3] P. Lecomte, D. Luckey, F. Nessi-Tedaldi, F. Pauss, Nuclear Instruments and Methods in Physics Research Section A 564 (2006) 164.
- [4] The CMS Electromagnetic Calorimeter Group, P. Adzic et al., JINST 5 P03010, 2010.
- [5] E. Auffray, M. Korjik, A. Singovski, On behalf of the CMS ECAL Group, in: Proceedings of SCINT 2011, 11th International Conference on Inorganic Scintillators and their Applications, Giessen, Germany and CMS Conference Report CMS CR 2011-257, to be published.
- [6] A. Singovski, E. Auffray, M. Korjik, On behalf of the CMS ECAL Group, Paper N29-4, IEEE/NSS 2011 Conference Record.

- [7] The CMS Collaboration, Technical Proposal for the Upgrade of the CMS Detector Through 2020, LHCC-P-004, CERN, Geneva, Switzerland, 2011.
- [8] F. Nessi-Tedaldi, *Journal of Physics: Conference Series* 160 (2009) 012013.
- [9] A.S. Iljinov, M.V. Mebel, C. Guaraldo, V. Lucherini, E. De Sanctis, N. Bianchi, P. Levi Sandri, V. Muccifora, E. Polli, A.R. Reolon, and P. Rossi, S. Lo Nigro, *Physical Review C* 39 (1989) 1420.
- [10] G. Dissertori, P. Lecomte, D. Luckey, F. Nessi-Tedaldi, F. Pauss, Th. Otto, S. Roesler, Ch. Urscheler, *Nuclear Instruments and Methods in Physics Research Section A* 622 (2010) 41.
- [11] M. Kobayashi, et al., *Nuclear Instruments and Methods in Physics Research Section A* 206 (1983) 107.
- [12] F. Nessi-Tedaldi, *International Journal of Modern Physics A* 20 (2005) 3826.
- [13] M. Korjik, in: *Proceedings of SCINT 2011, 11th International Conference on Inorganic Scintillators and Their Applications*, Giessen, Germany, to be published.
- [14] F. Dessauer, *Zeitschrift für Physik* 12 (1923) 38.
- [15] L.T. Chadderton, I.McC. Torrens, *Fission Damage in Crystals*, Methuen & Co. Ed., London, 1969.
- [16] P.D. Townsend, P.J. Chandler, L. Zhang, *Optical Effects of Ion Implantation*, Cambridge Univ. Press, Cambridge, 1994.
- [17] B.P. Price, R.M. Walker, *Journal of Geophysical Research* 68 (1963) 4847; R.L. Fleischer, B.P. Price, R.M. Walker, *Journal of Geophysical Research* 69 (1964) 4885.
- [18] M. Glaser, L. Durieu, F. Lemeilleur, M. Tavlet, C. Leroy, P. Roy, ROSE/RD48 Collaboration, *Nuclear Instruments and Methods in Physics Research Section A* 426 (1999) 72.
- [19] R.L. Fleischer, B.P. Price, R.M. Walker, *Nuclear Tracks in Solids: Principles and Applications*, University of California Press, Berkeley, USA, 1975 (and references therein).
- [20] Carl Zeiss AG, 8714 Feldbach, Switzerland <<http://www.zeiss.com>>.
- [21] Oxford Instruments, Abingdon, Oxfordshire, United Kingdom <<http://www.oxinst.com>>.
- [22] N. Tomašić, A. Gajović, V. Bermanec, D.S. Su, M. Rajić-Linarić, T. Ntaflos, R. Schlögl, *Physics and Chemistry of Minerals* 33 (2006) 145.
- [23] P. Tolédano, U. Bismayer, *Journal of Physics: Condensed Matter* 17 (2005) 6627.
- [24] D.L. Mills, *Journal of Applied Physics* 51 (1980) 5864.
- [25] A. Annenkov, E. Auffray, G. Drobychev, M. Korzhik, V. Kostyleva, O. Kovalev, P. Lecoq, V. Ligoun, O. Missevitch, R. Zouevski, *Nuclear Instruments and Methods in Physics Research Section A* 537 (2005) 173.