

An overview of results on scintillating crystals exposed to high hadron fluences: a long journey towards understanding

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Ingredients

$LT_0 = I_{out} / I_{in}$

Depending on the environment, crystals can be exposed to:

- Ionizing radiation levels
- Hadron fluxes

and one needs to worry about how these might affect the above

Ionising radiation effects in crystals

 ... are known almost as long as electric phenomena

T.J.Pearsall (1830), J. Royal Inst. 1 (1830) 77

 \rightarrow Electric sparks cause coloration to colorless Fluorite (probably using a Leyden jar)

E. Goldstein, Ann. der Phys und Chem. 54 (1895) 371

- \rightarrow Cathode rays cause coloration to salt, "blue halite"
- \rightarrow the coloration reaches a saturation level

M. Belar, S.B. Akad. Wiss. Wien, IIa, 132 (1923) 45

removal of coloration observed in Fluorite, at a speed depending on T

R.W.Pohl (1926), Z. Physik 39 (1938) 36

 \rightarrow Understanding the mechanism of coloration, concept of "Farbzentren" = color centers

Technical applications:

 \rightarrow screens for TV and radars, night vision, gem color "enhancement"

 J.H.Schulman, W.D. Compton, "Color Centers in Solids", Pergamon Press (1963)

Ionizing radiation effects in crystals for calorimetry

1) Appearance of radiation-induced absorption bands

- ◆ Typically narrow in energy ("color centers")
- ◆ Reduction of Light Transmission (LT)
- ◆ Possibly loss of uniformity in Light Output

alkali halides (BaF₂, CsI) damage related to oxygen contamination oxides (BGO, PbWO_{$_A$}) damage related to oxygen vacancies and impurities

These are due to point-like defects, where a [vacancy](http://en.wikipedia.org/wiki/Vacancy_defect) in the crystal is filled by an electron, which tends to absorb light

Wavelength (nm)

Color Center

Afterglow detected in PbWO4 as single photoelectron counting rate after irradiation

◆ Noise increase in detected Light - *energy equivalent contribution negligible e.g. for PbWO₄* in LHC experiments (R.Y. Zhu et al., NIM A376(1996) 319)

Characteristics of ionizing radiation effects

- 3) No damage in scintillation mechanism (demonstrated in BGO, BaF₂, CsI(Tl), PbWO₄)
	- \rightarrow changes can be monitored through a light-injection system

Recovery of damage at room temperature can occur: It depends on crystal type and, within one type, from growth parameters

What about high hadron fluences?

Some studies performed in the past on other calorimeter crystals, e.g.: BGO, M. Kobayashi et al., NIM 206 (1983) 107-117 CsI, M. Kobayashi et al., NIM A328 (1993) 501-505

Questions

- ◆ Is there a specific, possibly cumulative damage from hadrons?
- ◆ If so, what is its quantitative importance?
- ◆ Does it affect the light transmission only, and can it thus be "easily" monitored?
- ◆ Or else, does it alter the scintillation mechanism?

 \rightarrow Studies on PbWO₄

Questions

- ◆ Understanding of hadron effects observed in PbWO₄
- ◆ Complementary tests to confirm our qualitative understanding
- ◆ Performance of different crystal types
- ◆ Crystals particularly suited for superLHC

 \rightarrow Studies on CeF₃ \rightarrow Studies on LYSO

Particle fluences

Expected particle fluences $\text{[cm}^{-2}\text{]}$ for 2500 fb⁻¹ in 14 TeV pp collisions

- \triangle Barrel (η < 1.5): ~10¹² cm⁻² charged hadrons
- \triangle End Caps (1.5 < η < 3): up to ~10¹⁴ cm⁻² charged hadrons

Neutrons:

◆ Below 20 MeV, no effect besides ionizing dose, tested up to 10¹⁴ cm⁻²

R. Chipaux et al. Proc. Mat Res. Soc. 358 (1994) 481

◆ Above ~20 MeV, effects as for charged hadrons

Main crystals for HEP calorimetry

From the Review of Particle Properties, C.Amsler et al., Phys. Lett. B 667 (2008) 1, and earlier versions

e^- and π irradiations of PbWO₄ at IHEP Protvino

Irradiation studies between 1 and 60 rad/h up to 2 krad

V.Batarin et al, NIM A512 (2003) 488-505 V.Batarin et al, NIM A530 (2004) 286-292 V.Batarin et al., NIM A540 (2005) 131-139

- \bullet Behavior similar for e- and π -
- Damage appears to reach equilibrium at a dose-rate dependent level
- \bullet No indication of damage to scintillation mechanism from π irradiation

Caveat:Total absorbed dose expected at LHC not explored. Additional, specific, possibly cumulative damage from hadrons not excluded.

Super-intense hadron beam tests of PbWO₄ at IHEP Protvino

Mixed beam of charged hadrons, neutrons and γ with dose rates of 100 krad/h.

Constant flux.

Damage increases with accumulated dose

- ◆ Unlike purely ionizing radiation damage
- ◆ Hint towards an additional, cumulative, hadron-specific contribution

V.Batarin et al, NIM A512 (2003) 488-505

Effect on PbWO₄ Light Transmission by 20 GeV/c protons and ⁶⁰Co γ

M.Huhtinen, P.Lecomte, D.Luckey, F.Nessi-Tedaldi, F.Pauss, Nucl. Instr. Meth. A545 (2005) 63-87

- 1) Irradiation using 20 (24) GeV/c protons
	- \bullet Flux between 5x10¹¹ p/cm²/h and 10¹² p/cm²/h up to various fluences: crystals *a, b, c, d, h*
	- \triangle Flux of 10¹³ p/cm²/h up to various fluences: crystals *E, F, G*

factor 20 in rates

- 2) Complementary ${}^{60}Co$ γ irradiations
	- \bullet Dose rate of 1 kGy/h as in a flux of 10¹² p/cm²/h up to various total doses: crystals *t, u, v, w, x, y, z*

23 cm (25 X0) long crystals used, produced by the Bogoroditsk Techno-Chemical Plant. All crystals of production quality.

 \rightarrow Quantify damage through the induced absorption coefficient μ_{IND} in Longitudinal Transmission (LT):

Transmission changes in PbWO₄ for 20 GeV/c protons and ${}^{60}Co$ γ

M. Huhtinen, P.Lecomte, D.Luckey, F.Nessi-Tedaldi, F.Pauss, Nucl. Instr. Meth. A545 (2005) 63-87

 \rightarrow Changes in Light Transmission are qualitatively different from those (absorption bands) caused by γ radiation

 \rightarrow A band-edge shift^(*) is observed with proton-damage (left), unlike for γ -damage (right) (explanation likely to be disorder causing an Urbach-tail

 \rightarrow evaluate damage further at the peak-of-emission λ = 420 nm

Features of p-irradiated PbWO₄ crystals

LT recovery features in hadron-irradiated PbWO4

Features of p-irradiated PbWO₄ crystals

Green Laser light (543.5 nm) is shone through a p-irradiated crystal

scattering is observed!

a Polaroid filter reveals the green scattered light is polarised

Features of p-irradiated PbWO₄ crystals

Red Laser light (600-700 nm) is shone through a p-irradiated crystal

a Polaroid filter reveals the red scattered light is also polarised

Features of PbWO4 Transmission after p-irradiation

 \rightarrow $\mu_{\text{IND}}(\lambda)$ is qualitatively different between proton - and γ -irradiated crystals

In proton-irradiated crystals, Rayleigh-scattering behavior is observed:

i.e. scattering off "dipoles" with dimension $< \lambda$:

! λ-4 dependence (see crystal *a"*)

 \rightarrow scattered light completely polarized

Not observed for γ -irradiated crystals (see crystals v and y)

Proton and γ damage vs. fluence in PbWO₄

 \rightarrow tested up to p-fluence $\Phi_p = 5x10^{13}$ cm⁻²

- \rightarrow over 2 orders of magnitude in ϕ_p
- \rightarrow over a factor 20 in rates.

◆ Stable *HIND* component grows linearly with fluence: it is cumulative

◆ No flux dependence observed

quality (given by $\mu_{\textit{IND}}^{\gamma_{\textit{std}}}$ obtained from \rightarrow γ -induced damage saturates. Level depends on the initial crystal standard certification procedure)

Correlation between changes in LT and in Light Output in PbWO4

P.Lecomte, D.Luckey, F.Nessi-Tedaldi, F.Pauss, Nucl. Instr. Meth. A564 (2006) 164-168

Scintillation excited by cosmic muons, light output measured with 2262B Photomultiplier (bi-alkali photocatode)

 \triangle Correlation between μ_{IND} (420 nm) and Light Output loss for crystals irradiated with protons

Light Output loss for crystals irradiated with γ from a ⁶⁰Co source

Within the accuracy of the measurement, the two correlations are compatible

 \rightarrow No additional, hadron-specific damage to the scintillation mechanisms observed

Proton and γ damage in BGO

 \bullet BGO (Bi₄Ge₃O₁₂), used e.g. in L3 and BELLE

Data extracted from M. Kobayashi et al., NIM 206 (1983) 107-117

◆ Band-edge shift present for proton-irradiation, which does not recover with time

 \bullet No band-edge shift in γ irradiations

◆ Qualitative behavior of proton damage similar to the one in PbWO4

◆ Proton damage behavior compatible with a linear dependence on proton fluence

Remark: in CsI, hadron effects totally swamped by the ionizing radiation damage

Understanding hadron damage mechanisms in PbWO4 and BGO

M.Huhtinen, P.Lecomte, D.Luckey, F.Nessi-Tedaldi, F.Pauss, Nucl.Instr.Meth.A545 (2005) 63-87

Specific features of proton damage:

- ◆ It is cumulative: it grows linearly with fluence
- ◆ It only affects Light Transmission, and can thus be monitored
- ◆ The scintillation mechanism is not altered
- \bullet It has a Rayleigh-scattering behavior = scattering off "dipoles" with dimension $\lt \lambda$

Consistent with:

Fission of Pb, W, Bi above a \sim 20 MeV threshold, with production of heavy breakup fragments

- \rightarrow range \leq 10 µm
- \rightarrow E \le 100 MeV
- \rightarrow dE/dx = $\mathcal{O}(10000 \times dE/dx \text{ (mip)})$

Along their tracks, the crystal structure is changed permanently \rightarrow dipole-like regions where displacement, disorder, strain fields

 \rightarrow This feature should be absent for crystals made out of elements with Z<71 (*) A test of low-Z crystals should confirm this understanding of damage mechanisms \rightarrow Test CeF₃ and LYSO (*) *A.S.Iljinov et al.,Phys. Rev. C 39 (1989) 1420-1424*

A phenomenon studied long ago

Fr. Dessauer, Zeit. Physik 12 (1923) 38

 \rightarrow Concept of thermal spike when an incident ion comes to a stop in matter

J.A. Brinkman, J. Appl. Phys. 25 (1954) 961

 \rightarrow Concept of displacement spike

L.T. Chadderton, Nature 195 (1962) 987

 \rightarrow Experimental evidence for the displacement spike from its discontinuous nature

Tracks from 238U fission in muscovite mica

L.T. Chadderton, "Fission damage in crystals" (1969)

 ! *"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 centres or dislocations remain"*

R.L.Fleischer, R.M. Walker, P.B.Price, "Nuclear Tracks in solids" (1975):

 \rightarrow Dating based on fission track counting in crystals

The issue is still controversial

a) Damage due to highly-ionizing Pb- and W-fission fragments *(disorder, displacement, "bulk damage")*

b) Frenkel-type defects *"The defect formed when an [atom](http://en.wikipedia.org/wiki/Atom) leaves its place in the lattice, creating a [vacancy,](http://en.wikipedia.org/wiki/Vacancy_defect) and becomes [interstitial](http://en.wikipedia.org/wiki/Interstitial_defect) by lodging in a nearby location"*

A direct visualisation of fission tracks would settle this issue once and for all!

Comparative proton and pion damage study in PbWO4

P.Lecomte, D.Luckey, F.Nessi-Tedaldi, F.Pauss, D.Renker, Nucl. Instr. Meth. A587 (2008) 266-271

Crystal W, tested with γ in May 2004, was cut into 3 sections, W1, W2 and W3, each 7.5 cm (8.4 X_0) long, and the prior γ damage was annealed by heating.

Three sections (W1, W2 and W3) of the same crystal, each 7.5 cm $(8.4 X_0)$ long

- **W1** and W3 irradiated with 24 GeV/c protons up to $\phi_p = (1.17 \pm 0.11)x10^{13} p/cm^2$
- **W2** irradiated with 290 MeV/c π^{+} up to $\phi_{\pi} = (5.67 \pm 0.46)x10^{13} \pi$ /cm²

at a flux
$$
\varphi_{\pi}
$$
 = 4.13 x 10¹¹ π /cm²/h

Light Transmission changes in PbWO4: pions versus protons

 \rightarrow LT shape similar after p and π irradiations

 \rightarrow Band edge shift present after π irradiation as well

Same change in Light Transmission shape after p and π irradiation

(magnitudes similar due to suitable choice of fluences)

Damage can be globally fitted as after proton-irradiation, with τ_1 = 17.2 days and τ_2 = 650 days

Francesca Nessi-Tedaldi, ETH-Zürich FNAL Research Techniques Seminar November 8, 2010

Comparative proton and pion damage study in PbWO4

 \rightarrow a prediction of damage has to take this into account

**inelastic hadronic interaction caused by a projectile above a given energy threshold*

Cerium Fluoride - a bit of history

Apologies for incomplete bibliography

After the pioneering work of understanding CeF_3 luminescence...

F.A. Kröger & J. Bakker, Physica VIII (1941) 628-646

and its rediscovery of its properties as a scintillator...

D.F. Anderson, IEEE TNS 36 (1989) 137-140 W.W. Moses & S.E. Derenzo, IEEE TNS 36 (1989) 173-176

It was subject to an intense research program and studies, mainly in the '90 ... Scintillation characteristics, production of long crystals, behavior in γ and MeV-neutron irradiations, matrix performance in particle beams, e.g.:

> *M. Kobayashi et al., NIM A 302 (1991) 443-446 Crystal Clear Coll., S.Anderson et al., NIM A 332 (1993) 373-394 R. Chipaux et al., NIM A 345 (1994) 440-444 E. Auffray, F. N.-T. et al., NIM A 378 (1996) 171-178 R. Novotny et al., NIM A 486 (2002) 131-135*

...as Cerium Fluoride was baseline in the CMS and L3P Letters of Intent.

CERN-LHCC-92-003 and CERN-LHCC-92-005

Cerium Fluoride history (contd.)

Summary of characteristics in

S. Majewski & C. Zorn, "Instr. in High Energy Physics", F. Sauli Ed., World Scientific (1993)

Cerium Fluoride was also considered for medical imaging applications

W.W.Moses, S. Derenzo et al., J. Lumin. 59 (1994) 89-100

Ability to grow crystals beyond 30 cm length was demonstrated

but R&D would have to be restarted on it, since no commercial production exists at present.

CeF3 is still used, e.g. for neutron capture cross-sections measurements!

Transmission electron microscopy picture of 10 µm CeF3 nanoparticles by S. Stange et al, Los Alamos, IEEE/NSS 2009

Cerium Fluoride p-irradiation study

G.Dissertori, P.Lecomte, D.Luckey, F.Nessi-Tedaldi, F.Pauss, T. Otto, S. Roesler, Ch. Urscheler, NIM A 622 (2010) 41-48

Apply same irradiation and measurements procedures used for PbWO4

- \rightarrow CeF₃:Ba crystal from Optovac from the '90s, 21 x 16 x 141 mm³ (8.4 X₀)
- \rightarrow First 24 GeV/c p-irradiation at the CERN-PS IRRAD1 facility, up to

$$
\Phi_p = (2.78 \pm 0.20) \times 10^{13} \text{ p/cm}^2
$$

followed by recovery measurements over more than 1 year

 \rightarrow Second 24 GeV/c p-irradiation up to

$$
\Phi_p = (2.12 \pm 0.15) \times 10^{14} \text{ p/cm}^2
$$

followed by mesurements over 1 year

 \rightarrow Transmission damage evaluated at λ , where peak of scintillation emission, for Ba-doping \sim 340 nm, according to:

> *W.W. Moses & S.E.Derenzo, IEEE TNS 36 (1989) 173-176 Crystal Clear Coll., S.Anderson et al., NIM A 332 (1993) 373-394*

$Cef₃$ Transmission changes with proton irradiation

important recovery over a few months

→ Nd³⁺ "dips", see e.g. Crystal Clear Collab., E.Auffray et al., NIM A383 (1996) 367-390

! band-edge drop is due to an allowed transition *(M.Schneegans NIM A344 (1994) 47-56)* thus remains very steep

 \rightarrow light transmission recovers for all λ , except for an absorption band that seems cumulative, sitting however where the emission drops off.

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\rightarrow evaluate damage further at the peak-of-emission \lambda = 340 nm
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CeF3 Light absorption after p irradiation

- Rayleigh scattering behavior, as observed for PbWO4 over most of the λ range, is absent

 \rightarrow this confirms that the dominant Rayleigh scattering observed in PbWO4 is linked to the production of highly ionizing heavy fragments

Remark:

 Nd^{3+} dips totally disappear when **HIND IS evaluated** \rightarrow not influenced by radiation \rightarrow no hidden bands underneath

- An absorption band is present at \sim 400 nm, away from the emission λ , not identified so far. The density of centers N x oscillator strength f calculated according to *D.L.Dexter Phys. Rev. 101 (1956) 48* for the 2nd irradiation is

N x f \sim 1.7 x 10 ¹³ cm⁻³

Recovery of CeF3 Transmission after p irradiation

- Crystal left in the dark, at room temperature. Periodic transmission measurements

From 1st irradiation:

 \rightarrow recovery time constants τ_1 =10 ± 2 days and τ_2 =70 \pm 9 days

 \rightarrow 90% of the damage observed at 18 d is recovered after 1 year

After 2nd irradiation, radioactivity allowed handling only starting after 2 months:

 \rightarrow Fix τ_1 =10 days, and fit τ_2 =70±9 days, compatible with 1st irradiation results.

 \rightarrow Track recovery further, whether complete

 \rightarrow The amplitudes and time constants of recovery indicate that at superLHC, hadron damage would never build up to a severe level in CeF3

LYSO - Lutetium Yttrium Orthosilicate

Cerium-doped silicate-based crystals were recently developed for medical applications.

LSO (Lu2SiO5:Ce) was first investigated as a phosphor

A.H.Gomes et al., Mat. Res. Bull. 4 (1969) 643.

then rediscovered as a promising scintillator and first grown in 1989

C. Melcher US Patent, No. 4958080, 1990

Mass-production was established for LSO

C. Melcher and J. Schweitzer, IEEE TNS 39 (1992) 502-505

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and for LYSO (Lu_{2(1-x)}Y_{2x}SiO_5:Ce)
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D.W. Cooke et al., J. Appl. Phys. 88 (2000) 7360-7362 T. Kimble et al., Proc. IEEE NSS 2002

Numerous studies of their characteristics have been performed: *R.H.Mao, L.Y.Zang and R.Y. Zhu, IEEE TNS 55 (2008) 1759 and refs. therein*

The performance under γ -irradiation was thoroughly investigated:

R.H.Mao, L.Y.Zang and R.Y. Zhu, IEEE TNS 54 (2007) 1319

The performance for precision calorimetry was studied:

 M. Thiel, W. M. Döring, V. Dormenev, P. Drexler, R. W. Novotny, M. Rost, A. Thomas, IEEE TNS 55 (2008)1425

Investigation of LYSO under proton-irradiation

G.Dissertori, P.Lecomte, D.Luckey, F.Nessi-Tedaldi, F.Pauss, IEEE/NSS Orlando 2009

 \rightarrow While no industrial mass-production for CeF₃ is presently set up, LYSO is being mass produced by several companies, since it is heavily used in high-precision Positron-Emission Tomography.

 \rightarrow LYSO has a very high light yield, and could thus perform adequately even with radiation losses

 \rightarrow y-radiation effects have been shown to be small, and dose rate dependent

 \rightarrow The capability to grow large ingots has been demonstrated. Drawback: Lutetium is rather expensive

Apply same irradiation and measurement procedures used for $PbWO₄$ and $CeF₃$

- \rightarrow LYSO:Ce crystal from SIC, 25 x 25 x 100 mm³ (8.8 X₀)
- \rightarrow 24 GeV/c p-irradiation up to $\Phi_p = (0.89 \pm 0.06) \times 10^{13} \text{ p/cm}^2$
- \rightarrow Peak of scintillation emission at \sim 430 nm, according to:

R.H.Mao, L.Y.Zang and R.Y. Zhu, IEEE TNS 55 (2008) 1759

LYSO Transmission changes with proton irradiation

The change in Transmission induced by p-irradiation at this fluence in LYSO is quite modest

 \rightarrow No recovery overall is observed between 20 days and 123 days after irradiation

 \rightarrow evaluate damage further at the peak-of-emission λ = 430 nm

LYSO Light absorption after p irradiation

 \rightarrow In LYSO, as in CeF3, Rayleigh scattering behavior, as observed for PbWO₄ over most of the λ range, is absent

this is a further confirmation that the dominant Rayleigh scattering observed in PbWO₄ is linked to the production of highly ionizing heavy fragments.

Such fragments -as anticipateddo not seem to be present in **LYSO**

Recovery of LYSO Transmission after p irradiation

 \rightarrow The evolution of the absorption induced by p-irradiation in LYSO is compatible with no recovery at all.

 \rightarrow A fit using one time constant could allow as much as 20% of the damage to recover with τ =14 ± 64 (!) days. Recovery will be tracked further...

 \rightarrow A second irradiation, at ~10 x fluence, will tell us whether there is any cumulative damage

Damage amplitudes versus p-fluence

 \rightarrow In PbWO₄ a fraction of the damage has a component with $\tau >> 1$ years : "permanent" Values do not change beyond 150 d. Damage is cumulative

 \rightarrow In CeF₃ damage recovers, thus choice of time after irradiation for comparisons arbitrary. Damage is not cumulative.

 \rightarrow In LYSO we observe no damage recovery so far. A second irradiation, at \sim 10 x fluence, will tell us whether there is any cumulative damage

Conclusions

- A hadron-specific, cumulative damage has been observed in PbWO4, which only affects light transmission. All characteristics of the damage are consistent with an intense local energy deposition from heavy Pb- and W-fission fragments and the strain fields they leave behind.
- Measurements of proton-induced absorption up to $\Phi_p = 2 \times 10^{14}$ p/cm² in CeF₃ show a damage which recovers at room temperature and is not cumulative
- Measurements of proton-induced absorption in LYSO show a damage which does not seem to recover at room-T, but is a factor 5 smaller than in PbWO₄ for $\Phi_p = 0.9x10^{13}$ p/cm². Proton irradiations will be performed at higher fluences
	- \rightarrow they should allow establishing whether the damage is cumulative in LYSO
- The absence of a dominant Rayleigh-scattering component in CeF₃ and LYSO confirms that in PbWO4 it is due to the large energy deposit of heavy fragments.
- Our measurements demonstrate that particularly resistant crystals exist, suitable for precision calorimetry in high fluences of energetic hadrons, as expected at superLHC

Hadron damage is not entirely about color centers. It is also about nuclear interactions and displacement of atoms!

For resistance to hadron damage one might want to consider low-Z materials!