Examensarbete

Study of Radiation Damage of Lead Tungstate Crystals in the Context of CMS ECAL

Daniel Edström
Examensarbetet utfört vid CERN
2011-04-07

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The Large Hadron Collider, LHC, at CERN is capable of producing proton-proton collisions at a total energy of 14 TeV. Four principal detectors are installed in the LHC to detect the various particles created in the collisions. In one of these detectors, the CMS, scintillating crystals of lead tungstate are utilized to convert the energy of electromagnetic particles to visible light. Due to the high amount of radiation in the detector, the performance of the crystals may degrade during the lifetime of the detector.

In this work, crystals such as the ones used in the CMS detector are irradiated with gamma radiation and protons to estimate the transmission loss in the crystals. The spontaneous recovery at room temperature is studied over a period of several weeks. Induced diffusion is also studied. In addition, techniques for reversing crystal damage are presented and studied. The techniques used are thermal bleaching, restoration by heating the crystals, and optical bleaching, restoration by exposing the crystals to light of a specified wavelength.

The light yield, the amount of light extracted per unit of deposited energy, for different crystals and different types of sources has also been compared. The sources used have been gamma, alpha and neutron sources.

The induced absorption of standard PWO endcaps at 422 nm after being exposed to 1 kGy gammas was measured to be in the range of 0.6 to 0.7 m$^{-1}$. For molybdenum doped crystals the corresponding value was measured in the range of 0.2 to 0.3 m$^{-1}$. When increasing the dose to 100 kGy the induced absorption at 422 nm was found in the range of 1.17 to 1.32 m$^{-1}$ for standard crystals and 3.02 to 4.43 m$^{-1}$ for molybdenum doped crystals. The transmission could be completely recovered using thermal bleaching for both gamma and proton induced damage, while optical bleaching has so far only been shown to perform a limited recovery. Proton irradiated samples demonstrated an induced diffusion, which could be eliminated using thermal bleaching.

The light yield of the measured crystals was shown to be consistently lower for alpha particles and neutrons than for gamma particles. The rates varied in the range of 5% to 27% when comparing the light yield for alpha particles to gamma particles, and in the range of 5% to 70% when comparing neutrons to gammas.
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Abstract
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**Sammanfattning**


I detta arbete har kristaller av den typ som används i CMS-detektorn bestrålats med gammapartiklar och protoner för att uppskatta förlusten av transmission i kristallerna. Den spontana återhämtningen av transmission har studerats över en period av flera veckor. Inducerad diffusion från protoner har också studerats. Tekniker för att återfå kristallernas egenskaper presenteras och deras effektivitet studeras. Teknikerna som används är termisk blekning, återhämtning genom uppvärmning av kristallerna, och optisk blekning, återhämtning genom att belysa kristallerna med ljus av en specifik våglängd.

Mängden ljus en kristall producerar per enhet av deponerad energi för olika kristaller och typer av källor har studerats. De använda källorna har varit gamma-, alfa- och neutronkällor.

Den inducerade absorptionen hos standard endcap-kristaller vid 422 nm efter 1 kGy gammastrålning uppmättes mellan 0,6 och 0,7 m⁻¹. För molybdén-dopade kristaller var det motsvarande värdet mellan 0,2 och 0,3 m⁻¹. Efter en dos av 100 kGy uppmättes den inducerade absorptionen mellan 1,17 och 1,32 m⁻¹ för standardkrystaller och mellan 3,02 och 4,43 m⁻¹ för molybdén-dopade kristaller. Transmissionen kunde helt återfå medelst termisk blekning för både gamma- och protoninducerad skada, medan optisk blekning hittills endast uppvisat delvis återhämtning. Protonbestrålade prov visade en inducerad diffusion, som kunde elimineras med termisk blekning.

Mängden ljus de studerade kristallerna producerar visades vara konsekvent lägre för alfa- och neutroner än för gammapartiklar. Kvoten mellan mängden ljus producerat av alfa- och gammapartiklar varierade mellan 5 % och 27 % för de olika kristallerna. Motsvarande kvot för ljus producerat av neutroner och gammapartiklar varierade mellan 5 % och 70 %.
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Daniel Edström

Linköping, February 2011
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List of Abbreviations

ALICE  A Large Ion Collider Experiment
ATLAS  A Toroidal LHC Apparatus
BGO    Bismuth germanate \((\text{Bi}_4\text{Ge}_3\text{O}_{12})\)
CERN   European Organisation for Nuclear Research
CMS    Compact Muon Solenoid
LHC    Large Hadron Collider
LHCb   Large Hadron Collider beauty
LHCf   Large Hadron Collider forward
LuYAP  Lutetium Yttrium Aluminium Perovskite \((\text{Lu}_{1-x}\text{Y}_x\text{AlO}_3)\)
LY     Light Yield
LYSO   Lutetium Yttrium OxyorthoSilicate \((\text{Lu}_{2(1-x)}\text{Y}_{2x}\text{SiO}_5)\)
PMT    PhotoMultiplier Tube
PWO    Lead Tungstate \((\text{PbWO}_4)\)
QE     Quantum Efficiency
TOTEM  TOTal Elastic and diffractive cross section Measurement
Chapter 1
Introduction

1.1 CERN
CERN, the European Organization for Nuclear Research, is the world’s largest centre for particle physics. It was founded in 1954 and is located on the border between France and Switzerland, near Geneva.

CERN is currently composed of 20 member states. Furthermore, several states and organizations, including Russia, Japan, and the U.S.A, participate as observers. The main purpose of CERN is to provide scientists with the necessary tools to study the basic constituents of matter and the forces binding them together. To be able to study these building blocks of our universe, particles are accelerated in large particle accelerators and then collided. CERN provides scientists from 580 institutes and universities all over the world with access to accelerators, detectors, and computer centres for data processing.

Achievements at CERN include the discovery of the W and Z bosons which confirmed the unification of the electromagnetic and weak interactions, the electroweak theory of the standard model. This discovery led to the 1984 Nobel Prize for Simon van der Meer and Carlo Rubbia. The World Wide Web was also first invented at CERN in 1989, in order to facilitate information sharing between scientists across the globe.

1.1.1 LHC – The Large Hadron Collider
The most recent project at CERN is a circular collider called the LHC, the Large Hadron Collider. This machine, located 50 to 175 meters beneath the outskirts of Geneva, possesses a circumference of 27 kilometers and will, at full capacity, provide collisions between two beams of protons or lead ions with energy of 7 TeV each. In order to reach these energies, particles are accelerated in two separate beam lines to a velocity of 99.99% of the speed of light. The beams are guided in the ring structure by superconducting magnets cooled to a temperature of 1.9 K. The two beam lines intersect to form a collision point at four different points in the tunnel. At each of these collision points a detector has been constructed to observe the collisions and study the particles created. These detectors are: CMS, ATLAS, LHCb, and ALICE. Two smaller detectors, TOTEM and LHCf, are located near the CMS and LHCf detectors respectively, and will study the particles that pass each other closely rather than collide head on.

The purpose of the LHC is to attempt to further our understanding of the universe. This includes searching for the Higgs boson, which may explain the origin of mass, investigating the nature of dark matter, searching for extra spatial dimensions, recreating the conditions of the first few seconds of the universe and the reason for the dominance of matter over antimatter.
1.1.2 The CMS Experiment

The CMS, Compact Muon Solenoid, Detector is located beneath the village of Cessy in France. More than 2000 scientists from 155 institutes in 37 countries work with the CMS experiment. The CMS detector is designed as a general purpose detector and will observe a wide variety of particles and phenomena. The detector is constructed in layers, where each layer is designed to detect a specific class of particles. Going from the center and outwards, the first layer of the CMS detector is the tracking system. The tracking system records the paths of charged particle, which reveal their momenta. The second layer is the ECAL, the electromagnetic calorimeter. Consisting of scintillating crystals, this segment measures the energies of electrons and photons. The third layer is the HCAL, the hadronic calorimeter, which measures the energies of hadrons. The only known particles that may pass through the HCAL are the muons and weak interacting particles such as neutrinos. Muons will be tracked in the next layer, a dedicated muon tracking chamber. The neutrinos themselves may not be detected, but their presence may be inferred by adding up the momenta of the detected particles and attributing any missing momenta to the neutrinos. (1)

1.2 Scope of this Thesis

This thesis aims to cover a final thesis project for a Master of Science in Applied Physics and Electrical Engineering at Linköping University at the Department of Physics, Chemistry and Biology. The work has been carried out at CERN in Meyrin, Geneva, under the supervision of Etienne Auffray Hillemanns. The work commenced the first of March, 2010, and ended the 21st of December, 2010.

1.2.1 Problem Description and Objectives

The CMS electromagnetic calorimeter consists of over 70 000 scintillating lead tungstate crystals. The crystals emit light when exposed to ionizing radiation. The amount of light emitted is proportional to the energy absorbed from the radiation. This relationship enables the crystals to function as particle detectors. During the operation of the LHC the crystals will be exposed to large amounts of radiation. The exposure will over time create damages in the crystal, resulting in induced absorption and the subsequent loss of crystal transmission and light output. In order to prepare for the years to come, it is important to understand what amount of damage can be expected to occur in the crystals and if it is possible to recover from it. The aim of this thesis is to study the effect of radiation damage on the same type of crystals as those used in the CMS electromagnetic calorimeter. Both gamma irradiation and proton irradiation are studied. Techniques to recover from radiation damage are also presented and their efficiency measured. As a supplementary study, the light yield for various crystals from different kinds of particles has been studied and compared.

1.2.2 Limitations

The radiation absorbed by the lead tungstate crystals in the CMS electromagnetic calorimeter is expected to take place over the span of ten years. It is of course not possible to irradiate the crystals studied in this thesis for the same amount of time, and so the crystals studied in this work are instead irradiated over the course of several days.
1.2.3 Outline of this Thesis
The CMS electromagnetic calorimeter which the lead tungstate crystals are used in is a large and complex technical system, and as such requires a detailed introduction to be understood. This introduction is presented in chapter two. This chapter also serves to detail the mechanisms of scintillation. Important properties of scintillators are presented and described. The production methods and scintillation properties of lead tungstate are also presented. Chapter three details the mechanisms of damage recovery in scintillating crystals and presents the measurements apparatuses used in the recovery study. Chapter four presents the result of the recovery study for different types of radiation and recovery treatments. Chapter five outlines the light yield measurements when using different sources. Finally, chapter six provides discussion and conclusion for the work presented.
Chapter 2
Background

2.1 The CMS Electromagnetic Calorimeter
The purpose of the CMS Electromagnetic Calorimeter (ECAL) is to measure the energies of electrons, positrons and photons created in particle collisions. It will also contribute to the measurement of hadrons and missing energies in collaboration with the Hadron Calorimeter. In the search for the Higgs Boson, the ECAL will be instrumental through the measurement of the two-photon decay mode for \( m_H \leq 150 \text{ GeV} \) and the measurement of the electrons and positrons from the decay of the W- and Z-bosons originating in the \( H \to ZZ^{(*)} \) and \( H \to WW \) decay chain for \( 140 \text{ GeV} \leq m_H \leq 700 \text{ GeV} \). Measuring these phenomena requires excellent energy resolution and angular resolution. Further requirements are a fast response time and high resistance to radiation, due to operating conditions of the LHC.

The ECAL is composed of a central barrel part enclosed by two endcap parts, one at each end of the barrel. The barrel contains 61200 lead tungstate (PbWO\(_4\)) crystals, and the endcaps 7324 crystals each. Lead tungstate was chosen because of its properties making the crystals well suited to the CMS environment. Lead tungstate has a short radiation length and small Moliere radius, which enables a compact calorimeter. Furthermore, it has a fast response and is relatively easy to produce. The barrel crystals are arranged in submodules of ten crystals each and placed in an alveolar structure. These submodules are then arranged in modules of 40 or 50 submodules. Three modules of 40 and one module of 50 submodules make up one supermodule of which there are a total of 36. The barrel crystals measure 230 mm in length and have a cross-section of around 22*22 mm\(^2\). The exact shape of the crystal varies with their position in the barrel. The endcap crystals are arranged in supercrystals of 25 crystals. Each endcap consists of 276 standard supercrystals in addition to 36 partial supercrystals. All crystals in the endcap have the same size and shape. They are 220 mm long, the rear face 30*30 mm\(^2\) and the front face 28.6*28.6 mm\(^2\).

2.1.1 Photodetectors
The photodetectors used in the ECAL have several requirements placed on them. First of all, the PbWO\(_4\) crystals have a relatively low light yield, so they must have a high gain. Furthermore, they must be able to withstand both the high magnetic field and harsh radiation environment of the detector. Finally, they must have a fast response. With these requirements in mind, silicon Avalanche Photodiodes (APDs) are used in the barrel and Vacuum Phototriodes (VPTs) in the endcaps. For the barrel, two APDs are glued to the back of each crystal, covering 50 mm\(^2\) of the surface. The gain of the APDs is 50 and the quantum efficiency \( \approx 80\% \) for PWO crystals. The APDs cannot be used in the endcaps because they are not sufficiently radiation hard. Instead, VPTs are used. The gain of the VPTs is close to 10 and their quantum efficiency \( \approx 15\% \).
2.1.2 Preshower
The main purpose of the preshower detectors is to distinguish neutral pions from photons in the endcaps. They are situated in front of the endcap crystals and consist of layers of lead converters followed by detector planes of silicon strips. (3) In order to correct for the energy deposited in the lead converter, the energy measured in the silicon is used to apply corrections to the measurements in the crystals.

2.1.3 Radiation environment
The nominal luminosity of the LHC is $10^{34}$ cm$^{-2}$s$^{-1}$. Together with the beam energy of 7 TeV it will create a very hostile radiation environment. This can result in radiation damage in both the scintillating crystals and the electronic components. There is also the effect of induced radioactivity to be considered, which aside from being a safety issue may increase the level of background noise in the detectors. When discussing radiation, there are some units that need to be defined. Flux is the number of particles crossing a unit surface per unit of time. Fluence is the time integral of the flux; it is usually expressed in number of particles*cm$^{-2}$. Absorbed dose is the amount of energy deposited in a material per unit of mass. It is expressed in Gy. While particle fluence and absorbed dose are correlated for a given particle type, energy, and medium, they should not be treated as synonyms. (2)

Pseudorapidity is a spatial coordinate which describes the angle of a particle relative to the beam. It is defined as

$$\eta = -\ln\left(\frac{\tan(\theta/2)}{}\right)$$  \hspace{1cm} (2.1)

where $\theta$ is the angle relative to the beam.

Figure 1 shows the predicted dose profiles for the inner detectors of CMS for 500 fb$^{-1}$ and 2500 fb$^{-1}$ respectively. These values correspond to the end of year 2020 and 2028.

Figure 2 shows the predicted neutron and charged hadron fluence and absorbed dose immediately behind the crystals as a function of pseudorapidity. The values given correspond to 500 fb$^{-1}$. 
Figure 1: Predicted radiation profile in inner detectors. (4) The upper graph shows the predicted dose distribution for 500 fb$^{-1}$ and the lower graph for 2500 fb$^{-1}$. 
2.2 Scintillation
The defining property of a scintillator is the ability to absorb ionizing radiation (alpha, gamma, beta and X-ray) and reemit the energy in the form of visible light. Depending on the time taken for the emission, the scintillation can be divided into two categories. If the light is emitted with $10^{-8}$ seconds, which is roughly the time of atomic transitions, of the absorption the process is called fluorescence. If the process takes more time than this it is called phosphorescence, or afterglow. The time delay between the absorption and the emission is due to a metastable excitation state. Depending on the material, this delay can vary from a few microseconds to hours.

2.2.1 Scintillation mechanism
When a scintillator absorbs radiation it is excited into a non-equilibrium state. From this state, the scintillator will relax into equilibrium and emit light in the process. It is this process which is called scintillation. This relaxation occurs via several processes, but can be divided into five principal steps. (4)

1. The absorption of ionizing radiation causes the creation of primary holes and electrons.
2. The primary holes and electrons relax through the creation of secondary holes and electrons, photons, plasmons and other electric excitations.

3. Thermalization of low-energy electron-hole pairs results in a number of electron-hole pairs with energy just above the band gap energy \( E_g \).

4. Energy is transferred from the electron-hole pairs to the luminescent centres and their excitations.

5. The luminescent centres are deexcited via the emission of light.

The first three steps in this process are similar for all condensed matter. It is the second of these steps that is the most complex. The primary electron creates an electromagnetic cascade in the material consisting of electrons, holes, photons and plasmons. The number of these secondary particles increases with the depth of the cascade. The creation of secondary particles increases until the electromagnetic particles are unable to further ionize the material. At this stage the electron-hole pairs relax thermally to energy positions near the band gap energy. From this electromagnetic cascade some important parameters may be defined. One of these is the radiation length, \( X_0 \), which is defined as the mean distance travelled by an electron until its energy has been decreased by a factor 1/e. Another important parameter for scintillators is the Moliere radius, \( R_M \), which can be understood as the radius of an infinite cylinder containing 90% of the shower energy. The last two steps in the scintillation process are characteristic for different materials due to the involvement of the luminescent centres. These centres are the cause of the light emission and therefore most properties of the scintillation are caused by them.

### 2.2.2 Luminescent centres

The light emitted by a scintillator originates from radiative transitions between electronic levels which are called luminescent centres. The luminescent centres can either be intrinsic, meaning they originate from the crystal itself, or extrinsic, if their presence is due to impurities present in the crystal. In order to understand the mechanism of scintillation, we may employ the band theory. From this theory, we learn that the electrons of a completely free atom will occupy discrete energy levels given by Schrödinger’s equation. When atoms are brought together to form a solid, these energy levels will form a continuum of allowed energy levels, so called energy bands that the electrons may occupy. In between these bands are forbidden energy levels that the electrons may not occupy, so called energy gaps. In the normal state, the higher energy bands of the material are empty and the lower energy bands are filled with electrons. The filled band of the highest energy is called the valence band, and the empty band of the lowest energy is known as the conduction band. The valence band and the conduction band is separated by an energy gap known as the bandgap, \( E_g \). However, this model is only true for an insulator with a perfect crystal structure. In a real scintillator there are defects and impurities in the crystal lattice which give rise to local energy levels within the band gap which electrons may occupy. These levels may be characterized as three different types.

- Luminescent centres, from where the electron is relaxed via the emission of light.
- Quenching centres, from where the relaxation is achieved through thermal dissipation.
- Traps, a metastable level from where an electron may either gain energy from lattice vibrations and pass to the conduction band or lose energy and fall to the valence band.
The luminescent and quenching centres are caused by interstitial ions, impurities or defects. They introduce local energy states corresponding to the ground and excited state of the centre. Traps arise from other lattice defects and introduced energy levels below the conduction band.

![Energy diagram of a scintillator](image1)

Figure 3: Energy diagram of a scintillator. (5) The left picture shows the energy diagram of an insulator with a perfect crystal lattice. The right picture shows the energy diagram of a scintillator with quenching and luminescence centres and traps.

**Mechanism for the luminescence of a centre**

The mechanism of luminescence can be discussed using a theoretical model applicable to all luminescent materials. In this model, the potential energies of the ground and excited states of the luminescent centre are plotted against a configurational coordinate, the mean inter-atomic distance between the luminescent centre and its neighbouring atoms. Figure 4 is an example of such a plot.

![Potential energy diagram for a luminescence centre](image2)

Figure 4: Potential energy diagram for a luminescence centre. (5) The lines F1 and F2 correspond to the ground and excited states of the centre. The arrows A-C and B-D depict the excitation de-excitation processes.

In Figure 4 the minima A and B correspond to the stable energy positions of the ground and excited states, respectively. When the luminescence centre is excited, there is a transition from point A to point C. The transition is vertical in the configuration diagram because it is fast compared to the speed of atomic movements in the crystal. (3) In position C, the centre is not in its energy minimum and so it
moves to position B. The energy lost in the move is disposed of through thermal dissipation. The centre will remain in position B for a time that depends on the probability of an optical transition. This optical transition is what causes the emission of light. The optical transmission is vertical in the configuration diagram, again because of the speed of the emission in relation to the speed of atomic movements. Once in position D, the centre will move to position A by means of thermal dissipation. One may note that the emission occurs at lower energies than the absorption. This is what is known as Stokes’ Law, and the difference is expressed as the Stokes shift $\lambda_{\text{em}} - \lambda_{\text{ex}}$ where $\lambda_{\text{em}}$ is the wavelength of the emission and $\lambda_{\text{ex}}$ the wavelength of the excitation. A large Stokes shift indicates strong coupling between electrons and phonons. (6) The potential energy curves of the ground and excited states typically approach or intersect each other at some point. In Figure 4 this is point F. An excited luminescence centre in point F can make a non-radiative transition to point $F_1$ and from there dissipate the excess energy thermally. The probability of this process increases with temperature, as vibrational states closer to F may be occupied. This process is known as internal thermal quenching and competes with the scintillation process.

**Luminescence centres of Lead Tungstate**

PbWO$_4$ crystals have been shown to have three luminescent bands. (3)

- A blue band at 420 nm. This is present in all tungstates, which suggests it is caused by $WO_4^{2-}$ centres.
- A green band at 490 nm. When anion vacancies are present in the $WO_4^{2-}$ centres irregular anionic $WO_3$ molecular complexes form, causing this band.
- A red band at 650 nm. This band is also caused by irregular $WO_3$ centres, in this case deformed by Frenkel defects.

The luminescence spectrum of lead tungstate is the result of the combination of these three bands. However, the red and green centres are primarily related to slow components of the emission and are thus not desirable. For this reason these centres have been suppressed by means of special doping, resulting in an emission spectrum peaking at 420 nm.

**2.2.3 Scintillation properties**

**Light Yield**

The light yield is perhaps the most fundamental property of a scintillator. It is defined as the number of photons emitted by the scintillator per unit of deposited energy, typically MeV. When measuring the light yield, gamma sources are typically used.

$$LY = \frac{n_{\text{ph}}}{E_\gamma}$$

(2.2)

In this equation, $n_{\text{ph}}$ is the number of emitted photons and $E_\gamma$ is the energy of an incident gamma particle. The number of photons can be rewritten as

$$n_{\text{ph}} = n_{e-h}SQ, \quad 0 < S, Q < 1$$

(2.3)
Here, \( n_{e-h} \) is the number of electron-hole pairs created by the incident radiation. \( S \) is the efficiency of the energy transfer from the electron-hole pairs to the luminescence centres and \( Q \) is the efficiency of the energy transfer from the luminescence centre to emitted light. The energy required to create an electron-hole pair can be written as

\[
E_{e-h} = BE_g
\]  

(2.4)

where \( E_g \) is the bandgap and \( B \) is a material constant. Utilizing this relation, the number of electron-hole pairs created may be written as

\[
n_{e-h} = \frac{E_r}{BE_g}
\]  

(2.5)

Inserting this into our original equation thus yields

\[
LY = \frac{SQ}{BE_g}
\]  

(2.6)

With this relation, we may estimate the maximum theoretical light yield of \( \text{PbWO}_4 \) by setting \( S=Q=1 \). The band gap of \( \text{PbWO}_4 \) is \( E_g = 4.5 \) eV and it is known that \( B \approx 7 \) for tungstates. Inserting these values yield an estimation of \( LY_{\text{max}} \approx 32000 \) ph/MeV. In reality, the observed light yield for these crystals is closer to \( LY \approx 100 \) ph/MeV. The large difference in the theoretical maximum and measured values is due to the strong thermal quenching present in \( \text{PbWO}_4 \), resulting in a \( Q \ll 1 \). (5)

### Decay time

A scintillator’s decay time describes the amount of light emitted after a certain time. It is defined as the time after which the amount of light emitted has decayed to \( 1/e \) of its initial value. The light intensity as a function of time can be expressed using a sum of exponential functions. (3)

\[
I(t) = \sum_{i=1}^{n} A_i e^{-\frac{t}{\tau_i}}
\]  

(2.7)

Here \( A_i \) and \( \tau_i \) are the initial intensity and decay time of component \( i \), respectively, and \( n \) the number of components. This model accurately describes the intensity when one component is used for each luminescence centre in the scintillator. From this model, we may also define the contribution from each component, \( C_i \), and the mean decay time, \( \tau_m \). The parameter \( C_i \) is usually given as a percentage.

\[
C_i = \frac{A_i\tau_i}{\sum_{j=1}^{n} A_j\tau_j}
\]  

(2.8)

\[
\tau_m = \frac{\sum_{i=1}^{n} A_i\tau_i^2}{\sum_{i=1}^{n} A_i\tau_i}
\]  

(2.9)
The decay time is related to the mean lifetime of the excited states and therefore inversely proportional to the probability of de-excitation. The de-excitation can be either radiative or non-radiative, yielding

$$\tau \propto \frac{1}{P_r + P_{nr}}$$

(2.10)

Similarly, we define the luminescence efficiency $\eta$ as the ratio between the probability of a radiative de-excitation and the total probability of de-excitation.

$$\eta \propto \frac{P_r}{P_r + P_{nr}}$$

(2.11)

Looking at these two equations, we see that the luminescence efficiency and the decay time are at odds. Increasing $P_{nr}$ leads to a decreased decay time, and thus a faster response, but also decreases the efficiency which leads to a weaker emission.

**Radiation Hardness**

Radiation damage will always occur when ionizing radiation interacts with scintillation crystals. The creation of damage is a complex process depending not only on the material but also on defects and impurities in the crystal. Defects such as this increase the probability of the formation of radiation induced colour centres or point defects within the crystal. Changes in optical or scintillation properties of a crystal can be caused during irradiation or after the accumulation of large doses. The properties of the crystal can be impaired in several ways. (4)

- Formation of colour centre absorption bands which absorb the photons emitted by the luminescence centres, causing a decrease of the optical transmission. This is especially harmful in large gamma detectors for high energy physics.
- Direct effect on the emission characteristics. The luminescence centres may be quenched because of valence changes or ion diffusion. There may also be a reduction of the amount of emitted light due to reduction of energy transfers from electrons or holes to luminescence centres. The carrier mobility is reduced by the radiation induced defects and traps.
- Created shallow traps increase the amount of afterglow.
- Heavy crystals irradiated by high energy hadrons display induced radioactivity which can influence the operation of electromagnetic calorimeters.

A large amount of research shows that a loss of transmission is the most significant effect of radiation damage. For most crystals an observed loss in light yield can be explained by loss of transmission rather than luminescence. For this reason, the transmission is the primary property studied in radiation hardness studies. (7) Certain crystals may recover their transmission with time. The recovery at room temperature is often very slow, however. By means of thermal bleaching at an elevated temperature it is possible to obtain almost complete recovery. The temperature and time required depend on the properties of the colour centres. Another, more subtle, way of recovering radiation damage is by optical bleaching. This technique entails illuminating the damaged crystal with UV or visible light. If this technique can be demonstrated to work for the lead tungstate crystals, this provides an important tool
to deal with radiation damage inside the actual detector by bleaching the crystals during the periods when the detector is not operating. If the wavelength and intensity of the light is carefully selected it should be possible to fully recover the crystal transmission.

### 2.2.4 Creation of colour centres

When a crystal is exposed to ionizing radiation, the electrons are lifted from the valence band to the conduction band where they are free to travel in the crystal. The holes created in the valence band when the electrons are excited may also migrate. If the electrons or holes encounter a crystalline defect they may be trapped. The most studied case is the case of an electron trapped by a negative ion vacancy, which may be considered as an electron bound to a positive centre. A system like this generally possesses a spectrum of energy levels. Excitation between these levels produces a series of optical absorption lines analogous to those found in an isolated atom. (8) These centres are known as F-centres, from the German word ‘Farbe’, colour. F-centres may be arranged in groups. Two neighbouring vacancies trapping two electrons constitute an M-centre. Three neighbouring vacancies trapping three electrons are known as an R-centre. One vacancy trapping two electrons is known as an F'-centre. Holes are generally trapped by other defects than vacancies. A V-centre consists of a hole trapped between two neighbouring negative ions. This centre possesses an absorption spectrum similar to that of Cl₂. The introduction of impurities surrounding an F-centre generates a new family of colour centres. The most well-known, the F₄-centre is the result of the substitution of one of the six positive ions closest to an F-centre for an impurity. The reduction of symmetry of the centre induces a new absorption spectrum. (8)

### 2.2.5 Definition of induced absorption

The irradiation of crystals creates colour centres which modify the absorption of the crystal. This effect is caused by the energy levels associated with the defects, which causes a supplementary absorption. We may thus write the absorption of the crystal, μ, as a sum of the intrinsic absorption, μᵢ, and the induced absorption created by the colour centres, μᵢnd. (7)

\[
\mu = \mu_i + \mu_{\text{ind}}
\]  

(2.12)

The transmission of the crystal after irradiation is given by

\[
T_{\text{irr}} = T_{\text{th}} e^{-\mu L}
\]

(2.13)

where \(T_{\text{th}}\) is the theoretical transmission.

\[
T_{\text{irr}} = T_{\text{th}} e^{-\mu_i L} e^{-\mu_{\text{ind}} L} \]

(2.14)

\[
T_{\text{irr}} = T_i e^{-\mu_{\text{ind}} L}
\]

(2.15)

where \(T_i\) is the initial transmission. From this expression we may deduce the expression for the induced absorption.
\[ \mu_{\text{ind}} = \frac{1}{L} \ln \left( \frac{T_i}{T_{\text{irr}}} \right) \]  
\text{(2.16)}

When studying the effect of radiation damage and its recovery, the induced absorption is the quantity studied.

### 2.2.6 Recovery study

A number of studies of the radiation hardness of different materials have revealed that the colour centres created by irradiation are not always stable. As time passes, a decrease of the induced absorption can be observed in some cases. This recovery can be achieved at room temperature, but it is facilitated by increased temperature. It is also possible to observe this recovery when the crystal is illuminated by light. In this case we speak of optical recovery. \text{(7)}

#### Examples of recovery mechanisms

For instance, consider the simple case of an F centre, an electron trapped in an anion gap. We let \( P \) be the probability of the release of the electron from the trap and \( n(t) \) the density of F centres at the moment \( t \). In the case of direct recombination, the evolution of the density of F centres, at a given temperature, is governed by the following differential equation \text{(7)}:

\[ -\frac{dn}{dt} = P \cdot n(t) \]  
\text{(2.17)}

The solution to this equation is

\[ n(t) = n_0 e^{-Pt} \]  
\text{(2.18)}

where \( n_0 \) is the initial density of created colour centres. Because the absorption associated with colour centres is proportional to their density, there will be an exponential decrease of the absorption caused by the colour centres. If there is not direct recombination of the electron, the differential equation is not of the first order, and the recovery process will be slower. It is also interesting to study how the recovery changes with temperature. The probability of recovery \( P \) of a centre is given by Boltzmann's law:

\[ P = A e^{-\frac{E}{kT}} \]  
\text{(2.19)}

where \( A \) is a proportionality constant and \( E \) is the thermal deactivation energy. The higher this energy is, the harder it is for an electron or hole to escape a trap. In this case the damage is stable at ambient temperature. It is necessary to heat the crystal to a temperature so that \( E < kT \) to observe a recovery. If the recovery with temperature is accompanied by an emission of light, it is known as thermoluminescence.
2.3 Lead Tungstate Crystals

2.3.1 Crystal Production
The lead tungstate crystals for the CMS ECAL are produced at two different locations: Bogoroditsk Techno-Chemical Plant (BTCP) in the Tula region, Russia, and Shanghai Institute of Ceramics (SIC) in Shanghai, China. They are grown from a mixture of equal amounts of lead oxide (PbO) and tungsten oxide (WO\textsubscript{3}). The crystals are grown using two different procedures. At BTCP the Czochralski method is used, and at SIC the Bridgman-Stockbarger method is used.

In the Czochralski method, named after its inventor Jan Czochralski, the raw materials are melted in a crucible. A seed crystal is then attached to a vertical arm so that it is just in contact with the surface of the melt. The arm then slowly pulls the seed upwards, causing the melt to crystallize at the phase boundary. By rotating the crystal slowly it is possible to avoid replicating inhomogeneities of the melt in the crystal. Because the temperature, pulling speed and rotational speed influences the properties of the crystal, it is important they are kept constant. (6) An advantage of this method is that if the seed has an oriented crystal structure the grown crystal will have the same.

In the Bridgman-Stockbarger the crystal is grown inside a sealed container, the ampoule. The ampoule is slowly lowered from a hot zone, where the materials are melted, into a cold zone where the crystallization takes place. In order to control the growing conditions the hot and cold zones are heated separately. (6) The advantage of this method is that nearly all of the raw materials are used to form the crystal. A drawback is that some of the material may adhere to the walls of the ampoule, causing stresses which reduce the crystal quality.

2.3.2 Optical Properties

Theoretical Transmission

Transmission describes how much of the light entering the crystal at one side exits at the other side. PbWO\textsubscript{4} is an optically anisotropic medium of uniaxial type. This means that inside the crystal there will be both an ordinary ray, travelling according to the laws of isotropic media, and an extraordinary ray which travels with a velocity depending on the angle between the crystal optical axis and the interface between the crystal and the surrounding air. Because of this, the material has two indexes of refraction, the ordinary and an extraordinary index. An estimation of the transmission can nevertheless be obtained by considering the crystal as an isotropic medium. The complex index of refraction is \(N=n-ik\). From this, we may define the reflectance and absorption coefficient of a material.

\[
 r(\lambda) = \left| \frac{N(\lambda) - 1}{N(\lambda) + 1} \right|^2 \quad (2.20)
\]
\[ \alpha(\lambda) = \frac{4\pi k(\lambda)}{\lambda} \]  

(2.21)

In general, the transmission of a material is a function of these parameters and the thickness \( d \) of the material.

\[ T(\lambda) = \frac{(1 - r)^2 e^{-ad}}{1 - r^2 e^{-2ad}} \]  

(2.22)

Assuming that a perfect crystal only suffers transmission losses from reflections, we set \( \alpha(\lambda) = 0 \) and as such \( k(\lambda) = 0 \). The consequences for our equations are as follows.

\[ T(\lambda) = \frac{(1 - r)^2}{1 - r^2} \]  

(2.23)

\[ r(\lambda) = \left| \frac{n(\lambda) - 1}{n(\lambda) + 1} \right|^2 \]  

(2.24)

In order to find the refractive index \( n(\lambda) \) it is possible to use Sellmeier’s law, as formulated below.

\[ n^2 - 1 = \sum_{i=1}^{m} \frac{n_{s_i}^2}{1 - (\lambda_{s_i}/\lambda)^2} \]  

(2.25)

where \( n_{s_i}^2 \) and \( \lambda_{s_i} \) are the intensities and wavelengths of the optical resonances in the material. For lead tungstate, in the range of 320-850 nm, it is adequate to use a constant term for short wavelength resonances and one resonance term for the ultraviolet range. Sellmeier’s law thus becomes

\[ n_{ord}^2 - 1 = n_{s_0}^2 + \frac{n_{s_1}^2}{1 - (\lambda_{s_1}/\lambda)^2} \]  

(2.26)

The appropriate values for the parameters are expressed in the table below. (3)

<table>
<thead>
<tr>
<th>( n_{s_0} )</th>
<th>( n_{s_1} )</th>
<th>( \lambda_{s_1} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.5821±0.0080</td>
<td>1.1062±0.0097</td>
<td>270.63±0.82 nm</td>
</tr>
</tbody>
</table>

Table 1: Sellmeier parameters for lead tungstate

Using these parameters and the above equation, \( n_{ord} \) may be calculated. This may then be inserted into the expression for reflectance, which may in turn be inserted into the expression for transmission.
Figure 5: Theoretical and measured transmission of lead tungstate as a function of wavelength. The circled line shows the calculated theoretical transmission for an isotropic crystal without absorption. The dotted line shows the measured transmission.

This calculation is valid for an isotropic crystal without absorption. As described earlier this assumption is false for lead tungstate. A measured transmission spectrum for lead tungstate is displayed in Figure 5, together with the calculated spectrum. As seen in the figure, the curves agree with each other for $\lambda \geq 450$. For $\lambda \leq 350$ nm there is, however, a large disagreement. The reason for this disagreement is the assumption that there is no absorption. Within the band model, light with energy above that of the band gap will be absorbed to lift an electron from the valence band to the conduction band. The band gap of lead tungstate is 4.5 eV, or 275 nm. (3) The theoretical curve agrees with this consideration. However, a real crystal will also display internal absorption, which mostly occurs in the UV range. This internal absorption is the reason for the shift of the band edge in the real spectrum compared to the theoretical model. It is important for a scintillator that the transmission is as high as possible at the position of the emission peak, so that the crystal does not reabsorb the light emitted by the scintillation process. The emission spectrum of lead tungstate is shown together with the transmission in Figure 6.
Figure 6: Transmission and emission spectra of lead tungstate (3). The bold line shows the typical transmission spectrum for a lead tungstate crystal expressed as a percentage. The thin line shows the emission intensity in arbitrary units.
Chapter 3
Characterization of Crystals

3.1 Transmission bench – The Spectrophotometer
The purpose of the transmission bench is to compare the amount of light entering a crystal with the amount of light exiting it. This is done in order to find the light transmission as a function of wavelength. The main components of this transmission bench are a 150 W Xenon lamp, a monochromator, and a photomultiplier tube serving as a detector. The light emitted by the xenon lamp is focused at the monochromator and a set of filters located at the end of the monochromator serves to eliminate higher order diffractions.

![Schematic diagram of the spectrophotometer.](image)

The components of the spectrophotometer are as follows:

- A 150 W Xenon lamp emitting in the range of 200 to 700 nm.
- Several ultraviolet lenses to focus the light emitted by the lamp into the monochromator, and to collimate the light exiting the monochromator.
• A monochromator to select a specific wavelength. A motor coupled to the monochromator provides for automatic scanning of a given wavelength region.
• A number of filters to eliminate higher order diffractions from the monochromator. A stepping motor selects the filter to be used depending on the wavelength of the monochromator.
• A beam splitter that separates the light beam into two different beams, one traversing the crystal that is measured and one used as a reference.
• A chopper to alternate between the two beams. In order to make a correct measurement, the chopper must rotate with a frequency between 11 and 21 Hz. Should the chopper frequency drift outside of this range, an error is produced.
• A white box to defuse the light and make it arrive uniformly at the photomultiplier. This is because the measured signal depends on the impact position of the light on the photomultiplier cathode.
• A photomultiplier that measures the incident light.
• A moving table driven by a motor. Using this table several crystals can be measured without manually changing the crystal between each measurement.
• A LabVIEW program that controls the different parts of the machine and calculates the transmission coefficient from the measured light.

When performing a measurement, the signal from the photomultiplier is measured 25 times for each beam for every wavelength. From this data, the LabVIEW program calculates $K$, defined as

$$K(\lambda) = \frac{<M>}{<R>}$$

(3.1)

Here, $<M>$ is the average signal from the crystal beam, the beam passing through the crystal, and $<R>$ the average signal from the reference beam. This value does not depend on variations of the lamp or the photomultiplier. However, because the mirror reflectivity and beam splitter separations are functions of lambda, there will be some differences between the crystal beam and the reference beam. To compensate for this, a reference measurement without a crystal is performed every morning. This measurement is used to separate the signal differences caused by properties of the crystal being measured from those induced by wavelength dependencies of the equipment. For a reference measurement, the above equation becomes as follows:

$$K_{\text{ref}} = \frac{<M_{\text{ref}}>}{<R_{\text{ref}}>}

(3.2)$$

The final transmission can then be written as

$$T(\lambda) = \frac{K(\lambda)}{K_{\text{ref}} (\lambda)}$$

(3.3)
Because fluctuations in the power supply, lamp, detector and other irregularities are automatically compensated, these measurements are very stable. (3)

3.2 ACCOS
ACCOS stands for Automatic Crystal Control System. This system enables measurement of several crystals with minimal manual operation. The specific machine used for this work is called ACCoCE 2 and is installed at CERN. Up to 30 crystals can be loaded unto ACCoCE 2. This is done by means of a special loading system, so that the crystals need not be placed manually. The machine is capable of measuring longitudinal transmission, transversal transmission, light yield and the dimensions of a crystal. (3) Transversal transmission and light yield can be measured at several points along the length of the crystal.
Chapter 4
Results

4.1 Gamma irradiation

4.1.1 Comparison between PWO and PWMO
It has been speculated that doping PWO with molybdenum will increase its radiation hardness. To investigate this, both standard PWO crystals and molybdenum doped (PWMO) crystals have been irradiated with gamma rays.

Figure 8: Transmission of standard and molybdenum doped endcaps. The blue lines correspond to standard PWO crystals, while the red lines correspond to Mo-doped crystals.

Figure 8 shows the typical transmission spectra for two standard endcap crystals and three PWMO crystals. A distinct shift in the band edge in the PWMO crystals can be observed. This shift is in the order of 30 nm. Aside from this shift, the spectra are very similar. Scintillation properties are summarised in table 1 and Figure 9. The molybdenum doped crystals clearly have a higher light yield, at the cost of a slower response.
<table>
<thead>
<tr>
<th>Crystal ID</th>
<th>Ly at 8X0 (pe/MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Standard PWO Crystal</td>
<td>10.5</td>
</tr>
<tr>
<td>3011</td>
<td>15.3</td>
</tr>
<tr>
<td>3012</td>
<td>15.8</td>
</tr>
<tr>
<td>3013</td>
<td>15</td>
</tr>
<tr>
<td>3014</td>
<td>16.5</td>
</tr>
</tbody>
</table>

Table 2: Light yield of standard and molybdenum doped endcaps expressed in PE/MeV. Crystals 3011 to 3014 are doped with Mo.

Figure 9: Pulse shape of standard and molybdenum doped endcaps. The blue curve shows the scintillation pulse from a standard PWO crystal. The green curve shows the scintillation pulse from a Mo-doped PWO crystal. Measurement performed by E. Auffray.
Two standard PWO crystals and three molybdenum doped crystals were irradiated with 1 kGy gamma radiation, after which their transmission spectra were measured in ACCoCE 2 and the induced absorption calculated. Figure 10 shows the acquired induced absorption. It can be seen that the standard crystals display a higher induced absorption in the range 380-550 nm.

![Comparison PWO & PWMO Induced absorption after 1 kGy](image)

**Figure 10**: Induced absorption of standard and molybdenum doped endcaps after being exposed to 1 kGy gamma radiation.

The crystals were then heated so that their initial transmission was restored and then irradiated with 100 kGy gamma rays. The resulting induced absorption, along with the spectra obtained after 1 kGy, is displayed in Figure 12. The full lines are the induced absorption for the crystals after 100 kGy and the dash-dotted lines are after 1 kGy. In the case of 100 kGy the PWMO crystals display a much higher induced absorption. It can also be noted that the peak of the absorption is shifted towards the red end of the spectrum. Figure 11 shows the front face of the crystals as viewed with the naked eye. The left crystal is a standard PWO crystal and the right one a PWMO crystal.
Figure 11: Standard (left) and molybdenum doped (right) endcaps after 100 kGy irradiation

Figure 12: Induced absorption of standard and molybdenum doped endcaps after being exposed to 100 kGy gamma radiation. The dashed lines show the induced absorption after 1 kGy for comparison.

The crystals were then left in the ACCoCE 2 machine over a period of 85 days in order to study the recovery at room temperature. Figure 13 displays the measured induced absorption at the wavelength of 422 nm. The circles are measured absorption and the lines fits of an exponential and a constant term. The standard crystals do not display any notable recovery. However, a clear recovery can be seen in the absorption of the PWMO crystals.

A new PWMO crystal with a different amount of molybdenum doping was later received. The amount of doping was changed to improve the radiation hardness. This crystal measures 10 mm in length, and so it cannot be measured on the ACCoCE 2. It is therefore measured on the classical transmission bench.
Figure 14 shows the induced absorption after 100 kGy gammas of this crystal, PWMO 129, and of the previous endcaps, PWMO 3012. The induced absorption of the new crystal is in fact larger than that of the earlier endcaps. Figure 15 shows the recovery at room temperature for this crystal over a period of 34 days, measured on the transmission bench, along with a fit of an exponential and a constant.

![Graph showing induced absorption and recovery over time](image1.png)

Figure 13: Room temperature recovery of standard and molybdenum doped endcaps
Figure 14: Induced absorption of PWMO 129. The blue curve shows the induced absorption of PWMO 129 at 100 kGy. The green curve shows the induced absorption of crystal PWMO 3012 at the same dose, for comparison.
4.1.2 Thermal bleaching

After several days at room temperature, the absorption spectrum recovers very slowly or not at all. To accelerate the recuperation process, the crystals are heated. In effect, the colour centres of the crystals are destroyed by thermal activation. The oven used is a Nabertherm TR 60, equipped with a Nabertherm S 27 controller unit. The maximum temperature of the oven is 300ºC, and so this is the highest temperature at which we can study recovery. When the crystals are placed in the oven, the temperature is raised to the point that is being studied. The temperature is maintained for a predefined amount of time, after which the oven is allowed to cool. The crystals are taken out and measured on the transmission bench. Figure 16 shows evolution of the induced absorption at 420 nm for a standard endcap crystal and a molybdenum doped crystal. The time taken to bring the temperature to the specified point or cool the oven is not included in the time specified on the x-axis. There is limited recovery at 50 ºC, but a distinct recovery can be seen for temperatures of 100ºC and higher. At 200ºC there is complete recovery.
Figure 16: Thermal bleaching of standard and molybdenum doped endcaps.

4.1.3 Optical bleaching

An alternative possible way of recovery is to destroy the colour centre by illumination with light of a well chosen wavelength. A trapped electron is a system analogous to a hydrogen atom. It has different energy levels and can thus be excited by a ray of light. If the trapped electron acquires a high enough energy it can escape the trap, migrate in the crystal and recombine with a hole. To test this, prototype box with nine LEDs was produced, as seen in Figure 17. Two different sets of LEDs have been used. The first set is a set of diodes of the type B56L5111P from Roithner Lasertechnik, emitting at a wavelength of 475 nm. The second set uses diodes of the type ELD-740-524, also from Roithner Lasertechnik, emitting at a wavelength of 740 nm. The crystals are placed in front of the diodes so that the front face is illuminated. They are covered with black cloth to isolate them from other light sources. Figure 18 shows the induced absorption at 420 nm of a standard endcap crystal as well as a molybdenum doped crystal after treatment with optical bleaching. Some recovery can be observed at 475 nm, but none at 740 nm. In comparison to the thermal bleaching the effect is very small. The short molybdenum doped crystal was also treated with optical bleaching. The induced absorption at 420 nm for this crystal is displayed in Figure 19. There is little effect with the 475 nm diodes, but a notable effect with the 740 nm diodes.
Figure 17: Optical bleaching setup

Figure 18: Optical bleaching of standard and molybdenum doped endcaps
4.2 Proton irradiation

The effects of proton irradiation were studied on a number of PWO crystals measuring 10*10*20 mm$^2$ cut from a standard endcap crystal. They were first irradiated with gamma rays to compare the absorption spectra with those of the standard endcap crystals. Figure 20 and Figure 21 show the induced absorption of standard endcap crystals compared to the small crystals for 1 kGy and 100 kGy gammas respectively. Due to the shorter length of the small crystals, the measurements are more sensitive to fluctuations than those of the endcap crystals. However, the general shapes of the spectra are the same for endcap and small crystals. The small crystals were then irradiated with protons. Table 2 summarises the fluences and the rates the different crystals were exposed to. The induced absorption at 420 nm for the different crystal samples and the evolution at room temperature are displayed in Figure 22.
<table>
<thead>
<tr>
<th>Sample</th>
<th>Gammas</th>
<th>Protons</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.1</td>
<td>1kGy</td>
<td>$1.39E+13 , p/cm^2, 1.16E+13 , p/cm^2 , h$</td>
</tr>
<tr>
<td>1.2</td>
<td>1kGy</td>
<td>$1.51E+13 , p/cm^2, 1.33E+12 , p/cm^2 , h$</td>
</tr>
<tr>
<td>2.4</td>
<td>1kGy</td>
<td>$1.39E+13 , p/cm^2, 1.16E+13 , p/cm^2 , h$</td>
</tr>
<tr>
<td>3.1</td>
<td>100kGy</td>
<td>$1.51E+13 , p/cm^2, 1.33E+12 , p/cm^2 , h$</td>
</tr>
<tr>
<td>3.3</td>
<td></td>
<td>$1.64E+14 , p/cm^2, 1.3E+14 , p/cm^2 , h$</td>
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<tr>
<td>5.1</td>
<td></td>
<td>$4.53E+13 , p/cm^2, 5.5E+13 , p/cm^2 , h$</td>
</tr>
<tr>
<td>5.2</td>
<td></td>
<td>$2.26E+14 , p/cm^2, 4.6E+13 , p/cm^2 , h$</td>
</tr>
<tr>
<td>5.3</td>
<td></td>
<td>$2.26E+14 , p/cm^2, 4.6E+13 , p/cm^2 , h$</td>
</tr>
<tr>
<td>5.4</td>
<td></td>
<td>$8E+14 , p/cm^2, 5.6E+13 , p/cm^2 , h$</td>
</tr>
</tbody>
</table>

Table 3: Irradiation of small PWO samples. The centre column shows the gamma irradiation dose if performed. The rightmost column shows the flux and fluence of the proton irradiation.

Figure 20: Comparison of induced absorption after 1 kGy gamma irradiation of standard endcaps and small samples
Figure 21: Comparison of induced absorption after 100 kGy gamma radiation of standard endcap and small sample
Figure 22: Room temperature recovery of small samples of lead tungstate

4.2.1 Thermal Bleaching
Three of the small crystal samples were treated with thermal bleaching: 3.3, 5.2 and 5.4. However, the samples 5.2 and 5.4 were damaged and so only sample 3.3 has been studied over a full range of temperatures. Figure 23 shows the recovery of induced absorption at 420 nm for all three crystals with thermal annealing. There is little recovery at 50 ºC, followed by a notable recovery at 100 ºC. Figure 24 shows the recovery of sample 3.3 over the full range of temperatures. After several hundreds of hours at 300 ºC, all damage at 420 nm is recovered.
Figure 23: Thermal bleaching of proton irradiated samples
4.2.3 Optical Bleaching

One small sample, 5.3, has been treated with optical bleaching. The recovery at 420 nm is displayed in Figure 25. There is a notable recovery in the first 35 hours after which the absorption levels off. There is also some recovery when using the 740 nm diodes.
4.3 Diffusion

An effect that has been observed in proton irradiated crystal is the diffusion of light. To study this, a diffusion bench has been constructed. The bench consists of a laser, a beam splitter, a photodiode, a crystal holder and a camera. The beam splitter splits the beam from the laser in two beams, one that goes to a photodiode to measure the power of the laser and one that goes into the crystal. The camera is fixed in place above the crystal. In an ideal case, the beam going through the crystal should not be diffused at all and so nothing should be visible from the sides of the crystal. By taking a picture with the camera from above, the amount of diffusion can be measured. Figure 26 shows the pictures taken of series 5 before and after irradiation. From top to bottom they are sample 5.1, 5.2, 5.3 and 5.4. From left to right the pictures are before irradiation, top face after irradiation and side face after irradiation. There is a clear polarisation in the sample. This cannot be accurately quantified however, because the difference in light between the sides before irradiation was not measured. Figure 27 shows the difference in collected light from the top face for the different samples. The most irradiated samples are the ones displaying the largest amount of diffusion. Figure 28 shows the light collected from the top face of crystal 3.3 after finishing one temperature with thermal annealing, along with the amount of light collected. After treatment at 100 °C, there is a clear decrease in diffusion. The remaining diffusion likely pertains to mechanical surface damage rather than crystal defects.
Figure 26: Induced diffusion in proton irradiated samples. From top to bottom: Samples 5.1, 5.2, 5.3 and 5.4. From left to right: Before irradiation, top face after irradiation, side face after irradiation.

Figure 27: Difference in collected light before and after irradiation for samples 5.1, 5.2, 5.3 and 5.4.
4.4 Proton irradiation of standard endcaps

Twelve standard Endcap crystals were irradiated by protons at the PS facility IRRAD 3 at CERN. Ten of the crystals were produced at BTCP in Russia and two at SIC in China. The crystals were irradiated at a rate of $2 \times 10^{12}$ p/cm$^2$h, for a total fluence of $2 \times 10^{13}$ p/cm$^2$. Because of the high induced activation of the crystals, it is important to avoid manual manipulation of the crystals. For this reason, a special setup was constructed by the technicians of PH-CMX. A dedicated spectrophotometer was supplied by the MINSK group to allow for transmission measurements of the crystals without removing them from the room. The setup can be seen in Figure 29.

Figure 28: Recovery of diffusion with thermal bleaching in sample 3.3

Figure 29: Irradiation setup
Two of the irradiated crystals were left in the setup for the study of spontaneous recovery. The transmission of the other ten crystals was measured before and after irradiation. From this the induced absorption was calculated. The induced absorption of the crystals is displayed in Figure 30. The crystals with ID 4571 and 7022 respectively are the crystals produced by SIC.

![Induced absorption after proton irradiation](image)

**Figure 30**: Induced absorption of proton irradiated endcap crystals. Crystals 4571 and 7022 are produced by SIC, the others are produced by BTCP.

The ten crystals were placed in the center of a super crystal and taken to the test beam facility at H4 where the energy resolution would be measured and compared to that of non-irradiated crystals. This also allowed for the opportunity to test optical bleaching on site. Optical bleaching was tested on four of the proton irradiated crystal. After the test beam period, the crystals were again measured on the ACCoCE 2 machine. Figure 31 shows the removed induced absorption at 422 nm after the test beam period, where the removed induced absorption is defined as

\[
\text{Removed induced absorption} = 1 - \frac{\mu_{\text{ind,after test beam}}}{\mu_{\text{ind, before test beam}}} \quad (4.1)
\]

The four crystals with removed induced absorption around 20% correspond to the crystals which optical bleaching was tested on. Figure 32 shows the transmission before irradiation, before test beam and after test beam for one of these crystals.
Figure 31: Removed induced absorption after test beam period at 422 nm.

Figure 32: Transmission of crystal 11128 before irradiation, after irradiation and after bleaching in test beam.
Chapter 5
Comparison of Light Yield for Different Sources

Scintillating materials may react differently to different type of sources. Protons, α-particles and heavy ions are weakly penetrating particles. Because the light output of a scintillator not only depends on the energy losses of the incident particle but also on the density of ionization, the light output decreases. The consequence is that for equal energies, protons and α-particles produce less light than electrons. Neutrons are uncharged particles and do not ionize the material. The neutrons instead transfer energy to the atomic nuclei by means of the strong force. The scintillation is produced when the excited nuclei transfer to the ground state. The difference in light yield of different crystals depending on the type of incident particles has been investigated. The different sources are gamma, alpha and neutron sources. The crystals studied are LYSO, LuYAP, BGO, NaI, CsI, LaBr₃ and PWO.

5.1 Light Yield Measurements

In a light yield measurement, a radioactive source is used to initiate the scintillation process in the crystal that is being studied. The light produced by the crystal is detected by a photomultiplier tube. The sources that have been compared are two gamma sources, $^{60}$Co and $^{137}$Cs, an alpha source, $^{241}$Am, and a neutron source, AmBe. The crystal being measured is placed on the window of the photomultiplier tube with optical grease ($n≈1.5$) between the two surfaces. The purpose of the grease is to reduce the amount of light reflected at the interface between the media. The crystal sides are wrapped with reflective Teflon to prevent the loss of scintillation light through the sides of the crystal. As the light yield may vary with temperature a cooling unit is installed to maintain a constant temperature, typically 18ºC. Photons detected by a photomultiplier tube are absorbed by the photocathode. Through the photoelectric effect, the energy of the photons is transferred to an electric flux. The electron flux is multiplied through a series of dynodes before being collected by the anode and converted to an output signal. The efficiency with which the energy of the photons are converted to electrons is known as the Quantum Efficiency and is defined as follows:

$$QE = \frac{\text{Number of emitted photoelectrons}}{\text{Number of incident photons}}$$

(5.1)

This quantity generally varies with wavelength. Figure 33 shows a sketch of a photomultiplier tube.
5.1.1 Classical Light Yield Bench 4

The light yield measurements have been conducted on bench 4. The light produced by the crystal being measured is detected by a Photonis XP2262B photomultiplier tube. The photomultiplier tube is placed in a box which is covered by black cloth when a measurement is running to avoid any external light influencing the measurement. The bench is equipped with a Lauda RC 6 CS cooling system to maintain a stable temperature. The signal from the photomultiplier tube is collected by a LeCroy waveRunner 104Xi oscilloscope. Figure 34 shows a sketch of the electronics connecting the bench to the oscilloscope.
When a signal from the photomultiplier tube is detected a gate signal is generated. The signal from the photomultiplier tube is then integrated during the time the gate signal is high.

Figure 35: Scintillation pulse of LYSO

The part from 0 to 1 ns of the signal is also integrated and multiplied by five. This integral represents the electronic noise and provides the level of zero energy. Subtracting this value from the first integral gives the energy of the photoelectrons from the PMT. The values of the integral are stored and added to a histogram which results in a spectrum displaying the characteristics of the scintillation.

5.1.2 Extraction of Light Yield

To properly determine the light yield of a crystal, two measurements are performed. First, a measurement without any crystal or source is used. In this case the normal conduction electrons within the photocathode, which occasionally have enough energy to escape the photocathode, are detected. This results in a spectrum with a single peak representing the energy of a single electron. The second measurement is done with the crystal being studied and a radioactive source. This produces a spectrum representing the scintillation events in the studied crystal. Figure 36 shows a spectrum obtained from an LYSO crystal using a $^{137}$Cs source.
Some of the photons can undergo Compton scattering in the crystal. The photon interacts with an electron and is deflected at an angle $\theta$ with respect to its original direction. Considering the conservation of energy and momentum the following relationship can be derived. (4)

$$E'_\gamma = \frac{E_\gamma}{1 + \frac{E_\gamma}{m_e c^2} (1 - \cos(\theta))}$$  \hspace{1cm} (5.2)

In this equation, $E_\gamma$ is the energy of the incoming photon, $E'_\gamma$ is the energy of the photon after scattering, $\theta$ is the angle between the incoming and scattered photon, $m_e$ is the free electron mass and $c$ is the speed of light in vacuum. Scattered photons may leave the scintillator, in which case only the energy of the Compton scattered electron contributes to the scintillation light. The energy of the scattered electron varies with the angle of the scattering and reaches a minimum when the energy of the scattered photon is at a maximum. This occurs for $\theta=\pi$. The maximum energy transfer, $E_C$, can be derived from equation 5.2

$$E_C = E_\gamma \frac{1}{1 + \frac{m_e c^2}{2E_\gamma}}$$  \hspace{1cm} (5.3)
This is the absolute maximum of energy that a photon may transfer to an electron. This limit results in a sharp edge in the scintillation spectrum called the Compton edge. This is visible in Figure 36. The Backscattering peak, also visible in Figure 36, is caused by gamma rays scattered in the surrounding environment, such as the source shielding and crystal holder. The photopeak is the far right peak in the spectrum of Figure 36. This is the peak which characterizes the light yield of the scintillator. By fitting a Gaussian to the photopeak the position of the photopeak may be determined. The number of photoelectrons is found by dividing the position of the photopeak by the position of the single electron peak. The number of photoelectrons per MeV of energy is found according to the following equation.

\[
\frac{N_{pe}}{MeV} = \frac{\text{Position of photopeak}}{\text{Position of single electron peak}} \cdot \frac{A}{10^{20} E_s} (5.4)
\]

In this expression, A is the value of any applied attenuation in dB and \(E_s\) is the energy of the particles emitted by the radioactive source. To find the true light yield, this number needs to be divided by the quantum efficiency of the PMT. In this exercise, where we are comparing the light yield of a crystal for different sources, the quantum efficiency will be the same for all sources and it is sufficient to compare the number of photoelectrons.

5.1.3 Light Yield for Different Sources
Table 3 shows the particles emitted by the utilized sources. The energies and the relative intensities of the particles are included.

<table>
<thead>
<tr>
<th>Source</th>
<th>Co60 (9)</th>
<th>Cs137 (9)</th>
<th>Am241 (9)</th>
<th>AmBe (10)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gamma (E [keV,%])</td>
<td>1332.5, 99.99%</td>
<td>661.657, 85.1%</td>
<td>59.5412, 35.9%</td>
<td></td>
</tr>
<tr>
<td>Alpha (E [keV],%)</td>
<td>1173.2, 99.97%</td>
<td></td>
<td>5485.56, 84.5%</td>
<td>5442.80, 13.0%</td>
</tr>
<tr>
<td>Beta (E [keV, %])</td>
<td>318.13, 99.93%</td>
<td>513.97, 94.4%</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Neutron (E [keV])</td>
<td></td>
<td></td>
<td></td>
<td>~3000 ~5000 ~8000</td>
</tr>
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</table>

Table 4: Radioactive sources utilized for light yield measurements

Co60 and Cs137 are both gamma sources, and measurements done with these sources should thus result in the same light yield. The Co60 source displays two different energy peaks, and the spectrum acquired with this source should thus show two distinct photopeaks, as seen in Figure 37. However, in the case of low light yield crystals it may not be possible to accurately distinguish the two peaks. If this is the case, the mean energy of the gammas is used. The neutron source AmBe presents a similar case. For crystals with high light yield, three peaks can be observed. Figure 38 is an example of this. If only one peak is observed, the energy of the source is taken as 5 MeV.
Figure 37: Light yield spectrum of a LYSO crystal using a Co60 source

Figure 38: Light yield spectrum of an NaI crystal using an AmBe source
Table 4 summarizes the light yield obtained with the different sources. In the second far right column the quota of the light yields obtained with the Am241 and Cs137 sources is displayed and in the second far right column the quota of the light yield from the AmBe and Cs137 sources.

<table>
<thead>
<tr>
<th></th>
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<th></th>
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<td>LYSO</td>
<td>4150.668963</td>
<td>4555.593012</td>
<td>259.9321932</td>
<td>250.2387929</td>
<td>0.057057817</td>
<td>0.054930015</td>
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<td>4104.700314</td>
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<td></td>
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<tr>
<td>LUYAP</td>
<td>1589.680324</td>
<td>1699.296101</td>
<td>87.96807366</td>
<td>92.89167313</td>
<td>0.05176736</td>
<td>0.054664795</td>
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<td>1707.498005</td>
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<td></td>
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<tr>
<td>BGO</td>
<td>715.3370343</td>
<td>762.4879575</td>
<td>116.3136346</td>
<td>89.68348348</td>
<td>0.152544881</td>
<td>0.117619541</td>
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<td></td>
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<td></td>
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<td></td>
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<tr>
<td>NaI</td>
<td>10499.14618</td>
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<td></td>
<td>5835.471997</td>
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<td></td>
<td></td>
<td>4019.744001</td>
<td>0.265197232</td>
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<tr>
<td>CsI</td>
<td>8321.975833</td>
<td>8907.815911</td>
<td>56.06610024</td>
<td>5443.662078</td>
<td>0.006294034</td>
<td>0.611110752</td>
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<td></td>
<td>12298.87846</td>
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<td>3790.323953</td>
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<td></td>
<td></td>
<td>2685.075341</td>
<td>0.301429146</td>
<td></td>
</tr>
<tr>
<td>CsIpure</td>
<td>No Peak</td>
<td>83.71887429</td>
<td>22.42017758</td>
<td>58.74514159</td>
<td>0.267803142</td>
<td>0.701695312</td>
</tr>
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<td>LaBr3</td>
<td>7576.376038</td>
<td>11646.98719</td>
<td>1560.928573</td>
<td>4046.335176</td>
<td>0.134019944</td>
<td>0.347414753</td>
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<td>7356.52227</td>
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<td>2544.753245</td>
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<td></td>
<td>1647.902632</td>
<td>0.14148746</td>
<td></td>
</tr>
</tbody>
</table>

Table 5: Summary of light yield measurements

It can be seen that the calculated light yield for the AmBe source, in the case where there are three distinct peaks, differs for the three peaks. It is possible that the interpretation of the spectrum is wrong. The calculation of the number of photoelectrons produced versus the energy of the source for these cases is plotted in Figure 39. The equations of trendlines are summarized in Table 6.
Figure 39: Number of photoelectrons versus energy for NaI, CsI and LaBr3 using an AmBe source

<table>
<thead>
<tr>
<th>Crystal</th>
<th>Slope</th>
<th>Constant</th>
</tr>
</thead>
<tbody>
<tr>
<td>NaI</td>
<td>1266.2 PE/MeV</td>
<td>22262 PE</td>
</tr>
<tr>
<td>CsI</td>
<td>1015.2 PE/MeV</td>
<td>13507 PE</td>
</tr>
<tr>
<td>LaBr3</td>
<td>204.45 PE/MeV</td>
<td>11592 PE</td>
</tr>
</tbody>
</table>

Table 6: Trendlines for NaI, CsI and LaBr3 using an AmBe source
Chapter 6
Discussion and Conclusions

6.1 Irradiation damage and recovery

6.1.1. Gamma Irradiation
At a dose of 1 kGy, irradiated with gammas, the induced absorption for the standard PWO crystals PWO 11128 and PWO 11935 is 0.7032 m⁻¹ and 0.6007 m⁻¹ respectively at 422 nm. For the same dose and wavelength the induced absorption of the molybdenum doped crystals PWMO 3012, PWMO 3013 and PWMO 3014 is 0.3182 m⁻¹, 0.2013 m⁻¹ and 0.2216 m⁻¹ respectively. See Figure 10. When increasing the dose to 100 kGy the induced absorption for crystals PWO 11128 and 11935 at 422 nm is found to be 1.167 m⁻¹ and 1.32 m⁻¹ respectively. At the same wavelength and dose the induced absorption of the molybdenum doped crystals PWMO 3012 and PWMO 3013 is 4.432 m⁻¹ and 3.026 m⁻¹ respectively. See figure Figure 12. Molybdenum doped crystals thus seem to be more resistant to radiation damage at low dose rates, but perform worse at high doses. It can also be noted that the peak in the induced absorption is shifted towards higher wavelengths for the molybdenum doped crystals. For standard crystals a peak can be discerned in the vicinity of 400 nm. For molybdenum doped crystals, the maximum value is found in the vicinity of 600 nm. It is possible that the doping does in fact suppress the formation of the defects found in standard PWO crystals, but at high doses introduces a new kind of defect with an absorption centred on 600 nm. Leaving the crystals in room temperature and monitoring the induced absorption over time shows that the induced absorption at 422 nm of standard crystals remain at the same level. This can be seen in Figure 13. The induced absorption of the molybdenum doped crystals show an initial recovery, but remains larger than that seen in the standard crystals. Fitting the sum of an exponential and a constant gives a time constant of 32.9 days for PWMO 3012 and 32.2 days for PWMO 3013.

Crystals PWO 11128 and PWMO 3012 were treated with thermal bleaching. The results of the treatment are very similar for the two crystals. Figure 16, shows the induced absorption at 420 nm plotted versus the time spent in thermal bleaching for the two crystals. At 50°C there is a minor recovery, and when the temperature is increased to 100°C a strong recovery process can be observed. Fitting a sum of an exponential and a constant to this segment of the curve yields a time constant of 4.6751 hours for PWO 11128 and 4.0437 hours for PWMO 3012. When further increasing the temperature to 150°C the process is accelerated and after treatment at 200°C there is no remaining induced absorption.

Crystals PWO 11935 and PWMO 3013 where treated by means of optical bleaching, first with light of wavelength 475 nm and then with light of wavelength 740 nm. The induced absorption at 420 nm of these two crystals is shown in Figure 18. A slight recovery can be seen when using 475 nm light, particularly for crystal PWO 11935. At 740 nm there is little to no effect. Crystal sample PWMO 129 was also treated with optical bleaching, and the results can be seen in Figure 19. For this crystals there is little to no effect at 475 nm, but a clear recovery at 740 nm. Considering that the spectra of induced
absorption, shown in Figure 12 and Figure 14, showed a peak near 400 nm for standard crystals and a peak near 600 nm for the molybdenum doped crystals, it is likely that wavelengths closer to the peak of the absorption spectrum are more effective.

6.1.2 Proton Irradiation
A number of small samples cut from a standard endcap crystal were irradiated with various doses of protons. Of these, one sample has been treated with optical bleaching and three with thermal bleaching. Unfortunately, two of the samples treated with thermal bleaching where damaged during the treatment and so only partial data are available for these samples. Figure 23 show the induced absorption at 420 nm versus time spent in thermal bleaching of all three samples treated thermally. There is little to no recovery at 50 ºC, followed by a slight but noticeable recovery at 100 ºC. The complete recovery cycle for the undamaged sample, sample 3.3, can be seen in Figure 24. Complete recovery is achieved, but not until after several hundred of hours at 300 ºC.

Sample 5.3 was treated with optical bleaching and the results are shown in Figure 25. There is a clear recovery when using the 475 nm light, but it quickly reaches a plateau. When switching to the 740 nm light, there is some further limited recovery.

As seen in Figure 26, the proton irradiated samples display a clear diffusion. A polarization effect can be observed, however it cannot be accurately quantified because prior to the irradiation pictures were only taken of one side. A notable reduction in diffusion is observed when using thermal bleaching at 100 ºC. So far no reduction in diffusion using optical bleaching has been demonstrated. That the diffusion can be recovered using thermal bleaching suggests it is also caused by absorption centres, and not a more profound structural damage.

Ten standard encap crystals were irradiated with protons and measured in the test beam. Four of these were treated with optical bleaching during the test beam. The treated crystals display a reduction in induced absorption by 20%. The induced absorption of the non-treated crystals remains at the same levels.

6.2 Comparison of light yield for different sources
The light yield of crystal samples of LYSO, LUYAP, BGO, pure CsI, doped CsI and LaBr$_3$ was measured using different sources. The sources used were Cs137, Co60, Am241 and AmBe. Cs137 and Co60 are gamma sources, Am241 is an alpha source and AmBe is a neutron source. The measurements show that all measured crystals consistently produce less light when subjected to alpha radiation and neutrons than when subjected to gamma radiation. The fraction of light yields achieved with alpha radiation compared to gamma radiation ranges from 5% (LUYAP) to 27% (Undoped CsI). The fraction of light yields achieved with neutrons compared to gamma radiation ranges from 5% (LUYAP) to 70% (Undoped CsI).

NaI, doped CsI and LaBr$_3$ display three distinct peaks when measured with the neutron source (AmBe). It is known that AmBe possesses three emission peaks, and so it would be natural to assume that each emission peak is the source of one corresponding peak in the light yield spectrum. When using the energies of the emission peaks to calculate the light yield, however, each peak produces a different result, with the lowest energy peak giving the highest light yield. This work can produce no explanation
for why this is the case. One possibility is that the two extra peaks do not originate from the different emission peaks, but could perhaps be some form of backscattering or Compton edge.

6.3 Future work

In this work, recovery of proton irradiation damage has been limited to 10*10*20 mm$^3$ samples. It has been shown that thermal bleaching can fully recover the transmission of proton irradiated crystal. Optical bleaching has been shown to have an effect, but so far only a limited effect. In order to complete the study, tests must be performed on full size endcap crystals. Ten endcap crystals have been irradiated, and limited tests with optical bleaching have been performed in test beam. Complete tests with both thermal and optical bleaching should be performed for the proton irradiated endcap crystals just as they were for the gamma irradiated endcaps in this work.

The tests of optical bleaching in this work have been limited to two wavelengths, 475 and 740 nm. Using different wavelengths may yield a stronger recovery, and so it is interesting to test as many wavelengths as feasible. LEDs with emission peaks at 360 nm, 590 nm and 625 nm have been purchased.

In order to better understand the nature of the defects in the PWO crystals it is of interest to study the thermoluminescence of the crystals. This will require the construction of a dedicated thermoluminescence bench. This bench must be able to control the temperature and simultaneously measure light output and temperature. For this purpose a PID controller and thyristor unit has been purchased from Eurotherm.
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1. CMS - Detector. *CMS Experiment*. [Online] [Cited: December 18, 2010.]


