PROPERTIES AND RADIATION HARDNESS OF $PbWO_4$ CRYSTALS

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I. INTRODUCTION

Amongst new heavy scintillating crystals which satisfy the requirements of electromagnetic calorimetry in the extreme conditions of rate and radiation foreseen in the future projects as LHC, SSC, UNK single crystals based on lead and tungsten compounds have been already considered as a possible choice [1]. Recently it has been shown that $PbWO_4$ single crystals [2-6] possess such properties. The perspective of wide application of $PbWO_4$ scintillator for detectors has stimulated further investigations of these crystals to control the factors which determine its scintillation properties. Beam test have been performed at IHEP (Protvino) and at CERN.

II. BASIC PROPERTIES AND EXPERIMENTAL TECHNIQUES

The lead tungstate $PbWO_4$ single crystal structure is of the sheelite type (C^6_{4h} spatial group). It is isostructure to the well-known $PbMoO_4$ crystal [7] and, like $PbMoO_4$, it is birefringent along one axis. The main physical and chemical properties of $PbWO_4$ single crystal are summarized in the Table 1.

Table 1. Main $PbWO_4$ parameters

Dens. g/cm^3	Refr.ind. along Z axis	Melt. point°C	Hardn. (Moos)	X_0, cm	R_M, cm	Hygrosc.
8.28	2.16	1123	4	0.9	2.19	no

The investigated crystals were grown by the Czochralski method in platinum crucibles in an atmosphere close to air in composition. The initial mixture for crystal growth was prepared from PbO and WO_3 reagents with purity not less than 99.99%. The samples prepared for spectroscopic measurements were cut from the top part of the crystals which are usually grown to the dimensions : 32 mm diameter and 220 mm height. Absorption spectra have been measured with a BECKMAN UV 5760 spectrophotometer. The luminescence excitation spectra, photoluminescence and luminescence under the 122 keV γ-excitation of ^{57}Co have been measured with a SDL-2 spectrometer, and luminescence kinetics measured with a PRA 3000 spectrofluorimeter. A 500 R/s ^{60}Co source has been used to investigate radiation hardness. The scintillation kinetics have been measured by standard start-stop technique with a Lecroy QVT 3001 start by Phillips XP2262 PMT with an organic scintillator NE102A and stop by a PMT XP2020 detecting crystal light.

Beam test, with various samples of $PbWO_4$ crystals, have been performed at IHEP using 4, 9, 17 and 26 GeV electrons beams of $\sigma p/p \approx 2\%$ and at CERN with 10, 25, 50 and 100 GeV electron beams with $\sigma p/p$ of 0.4% or less. On both sites, a muon beam was used to measure the light produced by minimum ionizing particles and to measure the effective length of light absorption along the crystal.

III. OPTICAL SPECTROSCOPY

Single $PbWO_4$ crystals, polycrystals and samples obtained by a solid-phase synthesis have been investigated previously [7-14]. Unfortunatelly, yet there is no unique model of optical transition for radiating centers in $PbWO_4$ crystals that is in a good agreement with the collected experimental data. Luminescence characteristics of $PbWO_4$ single crystals are very sensitive to the peculiar conditions of their synthesis. Even small changes induce considerable variations in the emission and absorption spectra.

The results of the quoted papers may be summarized as :

1. In $PbWO_4$ single crystals, at least one band of emission is observed in the blue light range, two overlapping bands are observed in the yellow-green light range and two overlapping bands are observed in the red range.
2. All these bands are suppressed by strong temperature quenching.
3. The green luminescence line is polarized while the blue luminescence line is unpolarized.
4. There is no photoresistivity in crystals UV-excited near the edge of the intensive absorption band in the range with a wavelength $\lambda < 330$ nm, it only appears for $\lambda < 290$ nm.
5. The $PbWO_4$ samples exhibit fast decaying luminescence under pulsed X-ray excitation with a light yield of 460 photons/MeV.

By optimizing the conditions of $PbWO_4$ crystal growth the crystal coloration may be minimized and, light yield and scintillation efficiency have been increased and as well as the fast scintillation component has been enhanced.

The absorption spectrum is shown in Fig. 1. Optimal growth conditions may decrease the intensity of the 430 nm Pb^{3+} absorption band. Excitation in Pb^{3+} absorption band gives rise to a luminescent band with $\lambda_{max} = 650$ nm which is connected with the Pb^{3+} luminescent transition.

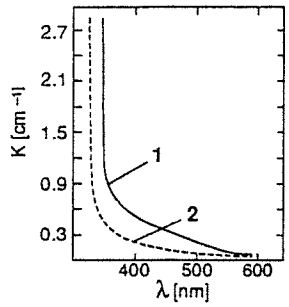

Fig. 1. Absorption spectra of $PbWO_4$ crystals grown in nonoptimal conditions (1) and after the technology optimization (2). T = 300°K.

The crystal excitation by light with λ less than 350 nm produces the luminescent bands with maxima at about 420, 490 and 650 nm. Fig. 2 shows the spectra of luminescence and excitation of luminescence. The detailed investigation of the excitation spectra has shown that the mean excitation peak λ_{ex} = 307 nm consists of two overlapping bands. Excitation from each wing of this peak generates two overlapping luminescence bands with close maxima, 495 and 481 nm. These results are in agreement with [10, 11]. Green luminescence bands are strongly polarized. The dependence of polarized ($\vec{E}$ // Z) luminescence intensity on the angle between Z crystal axis and polarisation direction of exciting radiation was measured with the technique described in [15].

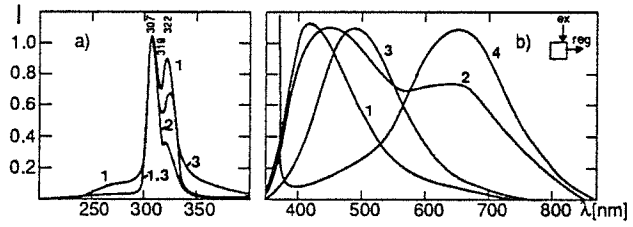

Fig. 2. Normalized spectra of the $PbWO_4$ luminescense excitation (a) : 1 - λ_{reg} = 430, 2 - 500, 3 - 600 nm and luminescence (b) : 1 - λ_{ex} = 275, 2 - 325, 3 - 308, 4 - 350 nm. T = 300°K.

Using these measurements, the C_3-like symmetry axes of green luminescence radiating dipoles have been determined. They have been shown to be very close to the diagonals of cube in which the WO_4^{2-} tetrahedron is inscribed. On this basis, we conclude that the green luminescent bands in the $PbWO_4$ crystals, in agreement with [9], are connected with the WO_3+ irregular centers. Moreover, these two green bands are caused by two different centers (WO_3+ F) similar to nonequivalent F-centers whose structure depends on oxygen vacancy between W^{6+} and Pb^{2+} ions or near W^{6+} ions.

The various luminescence bands have different kinetics under UV-excitation as shown in the Fig. 3. At room temperature the fastest one is the blue intrinsic luminescence originating from the regular WO_4^{2-} groups. The green luminescence bands have a similar decay component (out of 3), and the red luminescence is the slowest one with a decay time constant around 38 ns.

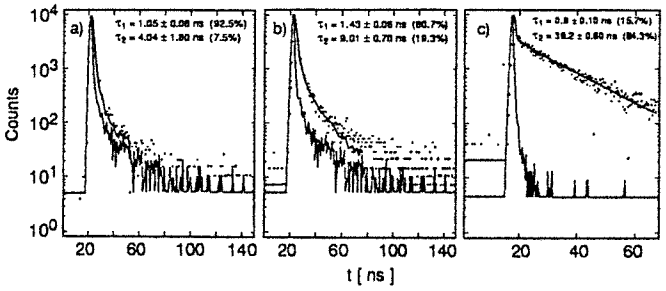

Fig. 3. Luminescence kinetics of $PbWO_4$ crystals.
a : λ_{ex} = 296 nm, λ_{lum} = 430 nm;
b : λ_{ex} = 296 nm, λ_{lum} = 490 nm;
c : λ_{ex} = 358 nm, λ_{lum} = 600 nm. T = 300°K.

Taking into account previous results of others authors, with our new measurements, we propose the energy level diagrams of the radiating centers in $PbWO_4$ shown in (Fig. 4).

The blue luminescence in $PbWO_4$ crystals is caused by the regular WO_4^{2-} groups, both green bands are connected with the irregular centers (WO_3+ F-center) and at least one of the red bands is generated by Pb^{3+} ions.

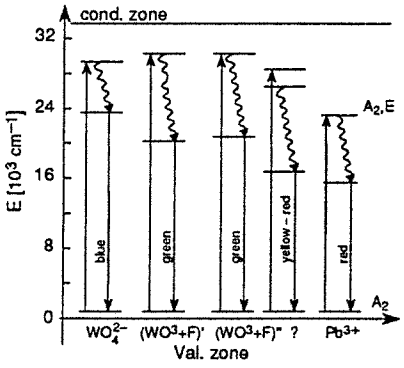

Fig. 4. Energy level diagrams of radiating centers in $PbWO_4$ crystals.

IV. SCINTILLATION PROPERTIES

Under the gamma-irradiation all $PbWO_4$ luminescent bands are excited and superposed. Crystals that are grown in optimal conditions show a spectrum with a maximum at λ ≈ 485 nm, with the FWHM of about 130 nm (Fig. 5).

The scintillation decay time has been measured. It depends on the maximum of luminescent band superposition spectra as shown on fig. 5 for different crystals. Various decay time components are reported in Table 2.

Table 2. $PbWO_4$ scintillation components

blue (420 nm), τ, ns (%)	blue-green (460 nm) τ, ns (%)	green (495 nm), τ, ns (%)
2.5 (> 98)	1.2 (28); 6.8 (43); 49 (29)	1.4 (32); 7.4 (39); 33 (29)

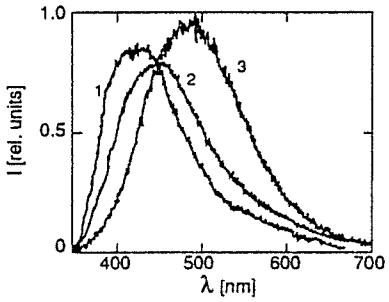

Fig. 5. Gamma-ray excited luminescence of $PbWO_4$ crystals grown in non-optimal (1,2) and optimal conditions (3). Eγ = 122 KeV (^{57}Co). T = 300°K.

The typical scintillation kinetics for crystals grown in optimal conditions is shown in Fig. 6. Slow components with the decay time of 80 ns and 337 ns are observed in these crystals, they represent less than 0.1% of the total light emitted.

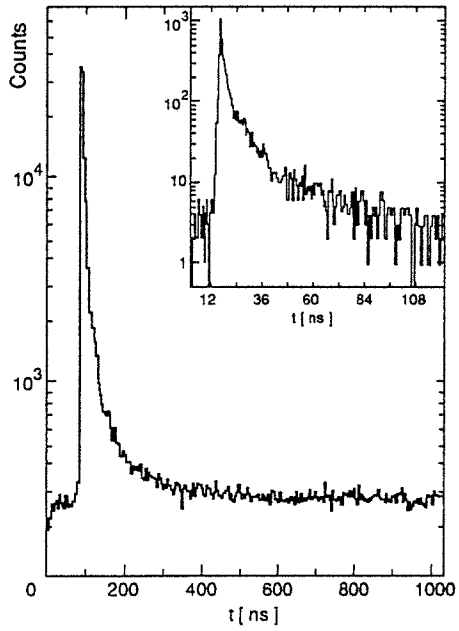

Fig. 6. Typical scintillation kinetics of the $PbWO_4$ crystals grown in optimal conditions. T = 300°K.

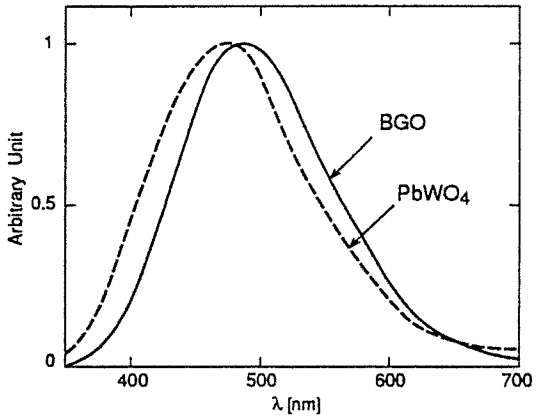

Fig. 7. The spectra of gamma-excited luminescence of $PbWO_4$ and BGO samples. Eγ = 122 KeV (^{57}Co). T = 300°K.

The comparison of $PbWO_4$ and BGO γ-ray excited luminescence is made in Fig. 7. Fig. 8 shows the amplitude spectra of $PbWO_4$ and BGO crystals excited by different sources. The scintillation yield of $PbWO_4$ crystals is about 5% relative to BGO, when bialkali photocathode PMT (XP-2262, XP-1911) is used as well as PMT FEU85.

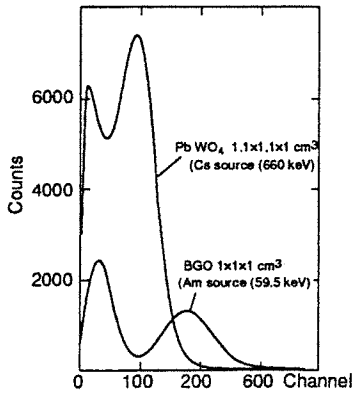

Fig. 8. Amplitude spectra produced in $PbWO_4$ and BGO scintillators by 600 KeV (^{137}Cs) and 60 KeV (^{241}Am) photons, respectively. T = 300°K.

V. RADIATION HARDNESS

The radiation hardness of $PbWO_4$ crystals depends on the condition of their growth. The transparency of coloured crystals increases under small irradiation doses (Fig. 9b) due to resorption of the 430 nm absorption band and latter decreases with larger doses. Similar effects have been observed by annealing crystals in vacuum and in air. It gives us arguments to correlate this effect with charge exchanges of $Pb^{3+} \rightarrow Pb^{2+}$ ions.

The $PbWO_4$ crystals recover their original transparency quickly after the irradiation, within ten days, the transparency is back very close to its original value before irradiation. The

crystals grown in optimal conditions show negligible changes of their transparency up to an absorbed dose of 5 Mrad (Fig. 9a).

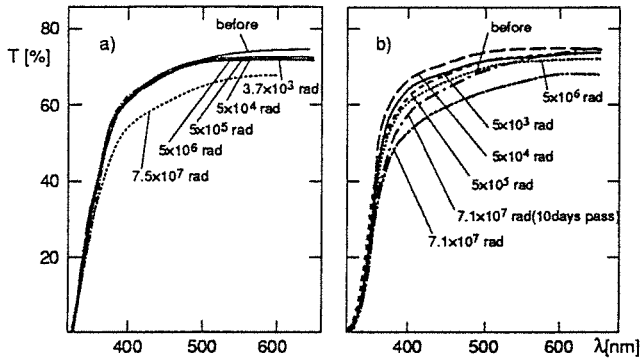

Fig. 9. Typical dependence of transmission of $PbWO_4$ samples (1 cm long) on the absorbed dose. a - crystals grown in optimal conditions, b - coloured crystals. T = 300°K.

VI. BEAM TEST

a) Electron beam tests

The beam tests of an electromagnetic EM-calorimeter prototype made of $PbW0_4$ crystals have been performed at CERN using the GAMS setup (NA12/2 experiment). Pure electron beams of 10, 25, 50 and 100 GeV, with the momentum spread $\sigma_p/p = 0.4$ or less was used. Impacts of electrons were measured within 1 mm by two-dimensional scintillation hodoscopes.

A matrix of 3 x 3 $PbWO_4$ crystals, 20 X_0 long each (20 x 20 x 180 mm^3) was studied to estimate the constant and stochastic terms of energy resolution. Each crystal was wrapped up with millipore and viewed by the Phillips XP 1911 PMT covering one half of the crystal exit area only. Optical coupling was done with Dow Corning Q2 - 3067 with a refractive index of 1.48.

Similar beam tests had been made previously at IHEP [17]. Only 4 $PbWO_4$ cells surrounded by $NaBi(WO_4)_2$ had been used that time to measure coordinate resolution and the EM-shower profile in the $PbWO_4$ EM-calorimeter. An upper limits for the energy resolution has been also obtained.

Fig. 10a displays the shower profile obtained during the IHEP tests in the 26 GeV electron beam, a silicon micro strip detector of 200 μ pitch being used for position measurement X from the crystal axis.

Fig. 10b shows the calibrated amplitudes in the $PbWO_4$ matrix cells when the beam hits 4 x 4 mm^2 area around the central crystal axis during CERN tests. The spectra of the calibrated amplitudes sum of all nine crystals, measured with 25 and 100 GeV electrons are displayed on Fig. 11.

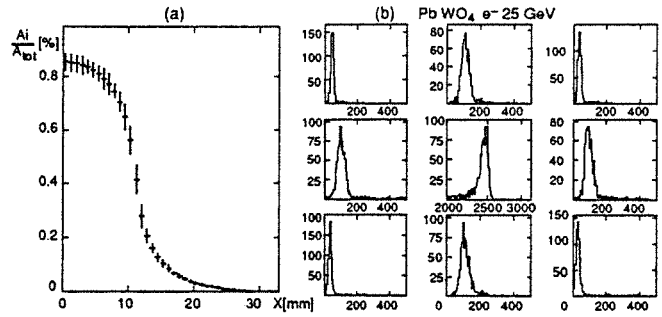

Fig. 10a. Shower profile in $PbWO_4$ measured with 26 GeV electrons.
Fig. 10b. Amplitudes spectra the $PbWO_4$ matrix produced by 25 GeV electrons hitting the central crystal (4 x 4 mm^2 beam spot).

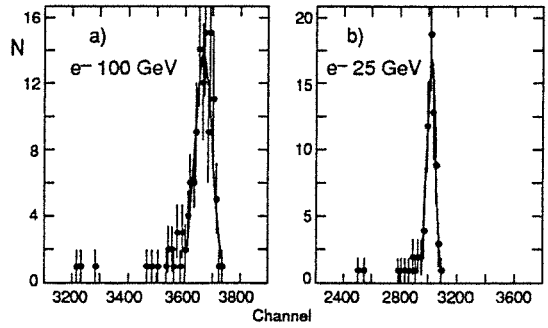

Fig. 11. $PbWO_4$ matrix amplitude sum obtained with the selected events (free of electronics drift).

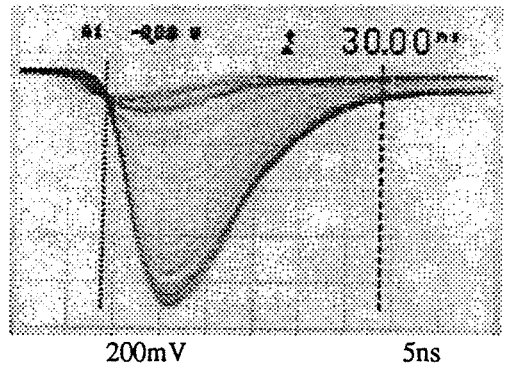

Fig. 12. Oscilloscope trace of 100 GeV electron shower in $PbWO_4$ rise time is 4ns.

They have σ_E/E of 0.96% and 0.81% respectively. Taking electron momentum spread $\sigma_p/p = 0.4\%$ into account, an energy resolution of $\sigma_E/E = 3\%/\sqrt{E} \oplus 0.64\%$ is achieved. A typical PMT signal observed with Tektronix 2467B oscilloscope with 100 GeV electrons after 4 m of fast 50Ω cable is shown on Fig. 12. The $PbWO_4$ crystals tested at CERN showed good timing characteristics. The fraction of the collected charge after 95 m of 50Ω cable is displayed versus the ADC gate width in Fig. 13.

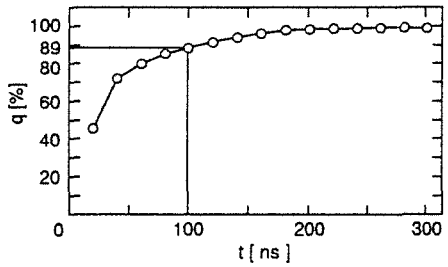

Fig. 13. Fraction of the collected charge versus, the ADC gate width. $\Delta t = 100$ ns is used during the beam tests.

b) Muon beam tests : The amplitude spectrum produced by muons traversing the $PbWO_4$ cell along its axis was measured with the HV settings used during the electron measurements. It is displayed in Fig. 14. The muon signal in the 180 mm long crystal corresponds to ≈ 0.3 GeV of EM-energy.

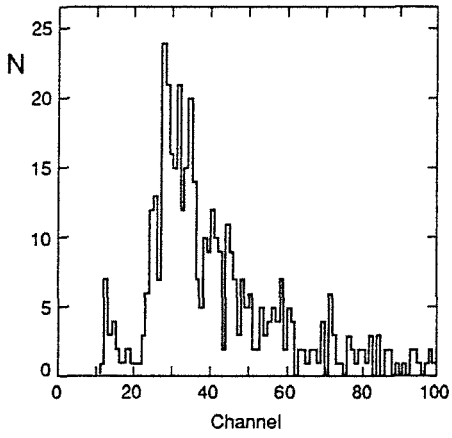

Fig. 14. Amplitude spectrum produced by muons crossing $PbWO_4$ cell along its axis.

The light produced by muons traversing the cell perpendicularly was measured in various points along its axis.. Results are shown in Fig. 15 for three different crystals. The uniformity of these crystals is high, the effective length scintillating light attenuation is around 2 m.

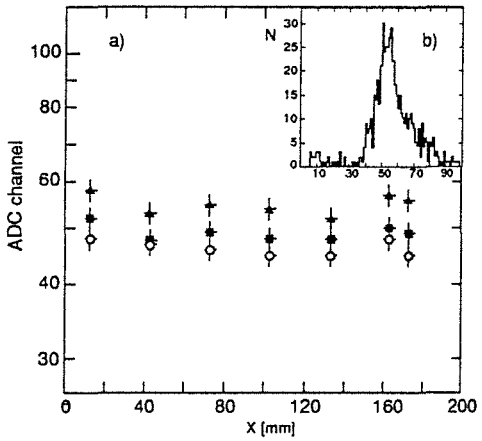

Fig. 15. Mean amplitude variation with the distance to PMT measured with muons crossing the cell perpendicularly to its axis. The insert shows the muon spectrum.

VII. ACKNOWLEDGEMENTS

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