FURTHER PROGRESS IN LEAD TUNGSTATE CRYSTALS

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Further Progress in Lead Tungstate Crystals

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I. INTRODUCTION

Recently, lead tungstate PbWO$_4$, also called PWO crystals, have been considered as a promising material for precise electromagnetic calorimetry [1–7] and the first tests have shown that energy resolution, with photomultiplier readout, better than 3%/ESR 0.7 can already be achieved with a calorimeter prototype of 9 or 20 counter cells (20×20×180+200 cm$^3$). Among the relevant properties of the crystal, its good radiation hardness has been specially mentioned as well as its short radiation length and its emission in the blue range of the spectrum. Further development in PWO technology to improve the transparency and uniformity of long crystals suitable for calorimetry has been undertaken. The usual growing conditions for normal PbWO$_4$ crystals have been tuned for a better control of stoichiometry, and investigation of pentavalent doping has been explored.

II. SPECTROSCOPIC AND SCINTILLATION PROPERTIES OF PWO SCINTILLATORS

A. Transparency and Growing Conditions

The improvement of PWO scintillation properties depends on the minimization of defects in the crystals based on Pb$^{1+}$ ions, caused by a shortage of cation in the W position [8]. As a result the transmission spectrum measured through a 20 mm thickness transversally to the growing axis of test crystal cells presents a difference between the seed side (also called top side) and the other extremity of the crystal (also called bottom side). This difference is shown on Fig. 1(a) for crystal grown in a limited-volume crucible of 100 mm in diameter.

A better uniformity of transmission along the crystal has been obtained by using a larger diameter crucible (120 mm) and using increased atmospheric pressure in the growing chamber. This is shown on Fig. 1(b) where no transmission spectrum difference has been observed between the top part and the bottom part of the crystal.

To minimize as much as possible the appearance of Pb$^{1+}$ defects or others, doping by Nb has also been used and found satisfactory [8]. Improvement of transparency along the Nb-doped crystal has been obtained as shown on Fig. 1(c). Compared with the crystal grown with the stoichiometry stabilization of Fig. 1(b), transparency in the 300–400 nm wavelength range has been significantly improved. Nevertheless, a weak absorption band appears near 420 nm.

![Fig. 1](image-url) Transmission curve through a 20 mm thickness of PWO crystal – top and bottom: a) grown in 100 mm diameter crucible; b) grown in 120 mm diameter crucible under increased atmospheric pressure in the chamber; c) grown with optimal Nb doping content.
B. Luminescence of Nb-Doped Crystal

To study the Nb doping effect on PWO crystal properties, a set of crystals containing different amounts of Nb has been investigated. All full-size crystals (with a length of at least 220 mm) were grown from the same crucible in the same conditions. The first crystal was grown without doping. For the other ones the Nb content expressed as a percentage of melt mass were 0.001 (N2), 0.0036 (N3), 0.0079 (N4), 0.015 (N5), 0.019 (N6), 0.022 (N7). The reference samples for detailed investigations with dimensions 10 × 10 × 10 or 20 × 20 × 10 mm³ were cut from the top of the grown ingot: for the N3 and N6 samples, pieces were also cut from the top and the bottom of the crystal. The distance between the top and the bottom samples cut from the same ingot is at least 190 mm.

![Fig. 2 γ-ray (17Co, 122 KeV) excited luminescence of the PWO:Nb samples N3, extracted from the top (a) and bottom (b) of the crystal. T = 300 K.](image)

The luminescence spectra excited by γ-rays are practically the same for the top and the bottom part of the crystal doped with a small amount of Nb. The luminescence spectra for the N3 sample are presented in Fig. 2 for the top and bottom parts. Although the Nb ions change the balance of the radiating centres in the crystal, the overall luminescence spectrum shape is not affected by the increasing proportion of (NbO₃ + F⁺) centres at the bottom part of the crystal.

III. DECAY TIME AND LIGHT YIELD OF Nb-DOPED CRYSTAL

A. Decay Time Measurement

The scintillation kinetics for Nb-doped PWO crystal samples has been measured by the usual start-stop method. Their decay time curves are well fitted by two exponential functions approximation.

\[ A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2) + A_3 \]

where A3 is the noise level of random coincidences.

The main parameters \( \tau_1 \) and \( \tau_2 \) of the fit are presented in Fig. 3 where N1 refers to the undoped crystal of the grown set. The slight decrease of \( \tau_1 \) and especially of \( \tau_2 \) with sample number is attributed to a decrease in the amount of the (NbO₃ + F⁺) centres amount the increase of the Nb content in the melt. So the scintillation of crystals with a significant amount of Nb is mainly produced by WO³⁻ centres and (NbO₃ + F⁺) centres with decay time constants 2.5 ns and 12 ns, respectively.

The difference between the kinetics of the top and bottom samples is small. The observed variation of decay time constants with increasing Nb content seems in good agreement with the scintillation mechanism expected from the model proposed elsewhere [8].

![Fig. 3 Decay time constant variation of Nb-doped sample (10 × 10 × 10 cm³) versus Nb content in the melt during crystal growth.](image)

B. Light Yield

The evaluation of light yield in photoelectrons/MeV has been obtained by comparison of the photopeak position of the 1.2 MeV γ-rays of ⁶⁰Co source measured by a Philips 2262 photomultiplier and the single electron peak of the phototube. The measurements have been done for reference samples as well as for 180 mm full-length crystals. For reference sample N3 (bottom part) 40 photoelectrons/MeV have been estimated. The effect of the Nb content in the reference samples on their light yield is summarized in Fig. 4 where the different samples are indicated on the horizontal scale referring to Nb content in mass per cent. Some improvement of light yield is observed for a 0.001 - 0.005% range of Nb content.
This can be due to the improved transmission below 450 nm of Nb-doped samples. Nb doping provides a better uniformity of light yield along the crystal.

Table I

<table>
<thead>
<tr>
<th>180 mm crystal</th>
<th>Top N\textsubscript{channel}</th>
<th>Bottom N\textsubscript{channel} (near PM window)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PWO undoped</td>
<td>160</td>
<td>190</td>
</tr>
<tr>
<td>PWO:Nb (0.0038%)</td>
<td>165</td>
<td>165</td>
</tr>
</tbody>
</table>

Table I gives the relative position (in channels) of the photopeak of the $^{60}$Co source for full size elements corresponding to N1 (undoped) and N3 samples, both 180 mm long, when the source is mounted at 1 cm (bottom) and 17 cm (top) from the photomultiplier window. The total light yield of Nb-doped crystal appears to be less than for undoped crystal when the source is near the PM window. A weak absorption band around 420 nm which was not present in the undoped crystal spectra appears and may give a small decrease of light yield. Its origin must be further investigated.

IV. RADIATION DAMAGE OF PWO:Nb-CRYSTALS

Radiation damage in PbWO$_4$ crystals is caused by charge exchange processes in the Pb$^{4+}$ structure point defects under $\gamma$-irradiation. An additional absorption band with a maximum around 620 nm appears for irradiated crystals with an intensity proportional to the absorbed dose [5, 9]. The crystal radiation damage near the seed (top part) is 10 times less than what is observed at the bottom part. With compensation of recharging defects, the radiation hardness of PbWO$_4$:Nb crystals is expected to be better.

A. $\gamma$-Ray Irradiation

Reference samples N1 (undoped), N3 and N6 (top and bottom) were irradiated by a 500 Rad's $^{60}$Co source up to a 0.5 Mrad absorbed dose. Transmission spectra, $\gamma$-ray excited luminescence spectra, scintillation kinetics as well as light yield were measured after irradiation (2-4 h). Transmission measurements are presented in Figs. 5 (a)-(e). PbWO$_4$:Nb crystals are seen as radiation hard even at low Nb content in the crystal.

Fig. 5 Changing of optical transmission of undoped (a) and Nb-doped samples extracted from the top (b, c) and the bottom (d, e) of crystals after $5 \times 10^2$ rad absorbed dose.

Measurements were done 2 h after irradiation.

From these data an estimation of the defects with Pb$^{4+}$ can be made. A compensation already exists for crystal N3 and defect concentration is estimated to be less than $5 \times 10^{17}$ cm$^{-3}$. A comparison of crystal characteristics before and after irradiation is given in Table II. A good stability of PWO:Nb crystal to $\gamma$-rays irradiation is observed. It indicates a negligible amount of defects recharged by $\gamma$-irradiation and structure point defects which could create colour centres in the crystal.

B. Electron Beam Irradiation

The full size PWO:Nb crystal corresponding to reference sample N3 was irradiated along its longer axis with a 500 MeV electron beam at the LIL facility at CERN. The impinging number of electrons corresponded to a 2 Mrad deposit at the maximum of the shower (referred to below as 2 Mrad equivalent). The effect of irradiation on crystal transmission is shown in Fig. 6 for different positions along the crystal from the top side. The homogeneity of the PWO:Nb crystal is indicated by the vicinity of the transparency curves.
Two months after irradiation the crystal had fully recovered its original transparency. Finally in Fig. 8 the absorption coefficient induced by irradiation is compared for an undoped crystal from a previous production batch measured in the RD18 collaboration at CERN after 0.076 Mrad irradiation by $^{60}$Co γ-rays and for a PWO:Nb crystal after 2 Mrad equivalent irradiation by the LLN electron beam. The doping with Nb further increased the good radiation hardness of normal PWO by decreasing the already small induced absorption coefficient ($\Delta K \sim 2.5 \text{ m}^{-1}$) by a factor 10 ($\Delta K \sim 0.25 \text{ m}^{-1}$) for Nb-doped crystal.

V. BEAM TEST FOR DIFFERENT CRYSTAL TYPES

A beam test was performed in the X1 beam at CERN for various types of PWO crystal [10]. Energy resolution and temperature dependence of the light yield was measured. The preliminary results of energy resolution with photomultiplier readout for an e⁺ beam between 10 and 70 GeV are presented in Fig. 9(a) for a matrix of 25 cells (20 × 20 × 180 + 200 cm$^3$) of PWO. It fits well the $\sigma E = 2.8 \sqrt{E} + 0.45$ formula where 0.3% of the beam momentum spread had been quadratically removed. The same performance is expected for an Nb-doped crystal matrix as can be seen in Fig. 9(b) where 20 GeV/c electron peak hitting a single cell within a 2 × 2 mm$^2$ area is shown for an Nb-doped and for an undoped cell. The temperature dependency of the light output of undoped PWO crystal is shown in Fig. 10 and found to be $-2.10 \pm 0.06\%$ °C$^{-1}$ at 17°C. Similar results have been obtained with Nb-doped crystals.
VI. CONCLUSION

The tuning of the growing conditions improved the uniformity of the stoichiometric PWO crystal. Crystals grown with Nb doping show improved transmission below 400 nm and, by controlling the occurrence of the defect in the crystal based on Pb$^+$ ions, their radiation hardness is increased significantly.

VII. ACKNOWLEDGEMENTS

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VIII. REFERENCES


PWO EM-calorimeter tests 1994

X1 beam
- matrix 5x8 with PMT readout
  - with 5 Nb doped crystals
- matrix 3x3 with EG&G APD
- matrix 3x3 with PIN diodes 1x1cm (Hemeros)
- matrix 3x3 with APD 10x10mm$^2$ from Minsk

Energy range 5 to 70 GeV, $\delta_{beam} \approx 0.3\%$

H4 beam
- matrix 5x5 with PMT readout
- uniformity measurements

Energy range 20 - 150 GeV

X1 beam
- CMS crystals, matrix 4x4
  - old wrapping
  - new - " - " -

Katchanov V. (Katchano@cernvm)
X1 beam tests.

CMS crystals, new wrapping
matrix 4x4

\[ N \]

20 GeV e⁻

\[ N \]

50 GeV e⁻

20 GeV: \( \frac{G}{E} \approx 0.87\% \)
50 GeV: \( \frac{G}{E} \approx 0.61\% \)

(without any corrections)

Beam spread is \( G \approx 0.3\% \)

with a beam correction

20 GeV 50 GeV

\( \frac{G}{E} \approx 0.81 \) \( \frac{G}{E} \approx 0.53 \)
PWO 5x5 matrix PM1911 readout 6x6 mm. e- beam

\[
\frac{\sigma E}{E} = 0.73\% \pm 0.45\% \pm 0.3\%
\]
X1 beam tests.

CMS crystals, new wrapping matrix 4x4

20 GeV: $\frac{E}{\theta} \approx 0.87\%$

50 GeV: $\frac{E}{\theta} \approx 0.61\%$

(without any corrections)

Beam spread is $\theta \approx 0.3\%$

with a beam correction:

20 GeV: $\frac{E}{\theta} \approx 0.81$

50 GeV: $\frac{E}{\theta} \approx 0.53$
ALICE/PWO
matrix 5x6

Pb NO{sub 4}, matrix 3x3
KEK 1cs1
K. Takamatsu

PMT zeadou

ENERGY RESOLUTION

\[ \frac{\sigma}{E} = 3.0\% \sqrt{E} + 0.88 \]

\[ \frac{\sigma}{E} = 2.65\% \sqrt{E} \pm 0.6 \]

- KEK data
- ALICE/PWO (no beam and leak corrections)

\( \text{Sigma/E} \)
\( \frac{1}{\sqrt{E}} \)

50 100 10 5.0 2.5 1.5 1.0 0.5
0 1 2

GEV
Linearity of PWO EM-calorimeter 23X0, from 5 to 20 GeV better than ±0.1%.
Coordinate resolution and lateral profile of EM-shower in PbWO$_4$

![Position resolution as a function of position](image1.png)

![Electromagnetic shower lateral profiles](image2.png)
PIN diode readout

MIP in crystal only 2.5 cm

\( \approx 25 \text{ MeV} \)

MIP in crystal and pipe

PIN diode readout - Ti60 and scint. light.

\( \approx 15 \text{ ph. MeV} \)

Corrective:
- quench: 2\% - 0.55
- scint. factor 4

\( e^- \quad 5 \text{ GeV} \)

\( 1 \text{ p} = 115 \text{ ph. MeV} \)

PWO light yield measurements
Noise of 9 cells

Noise for PIN diodes readout \( \sum_9 \approx 200 \text{ MeV} \)
PWO PM1911 readout. Light yield vs. temperature

\[ ( -1.99 \pm 0.01 ) \% / \text{deg.} \]