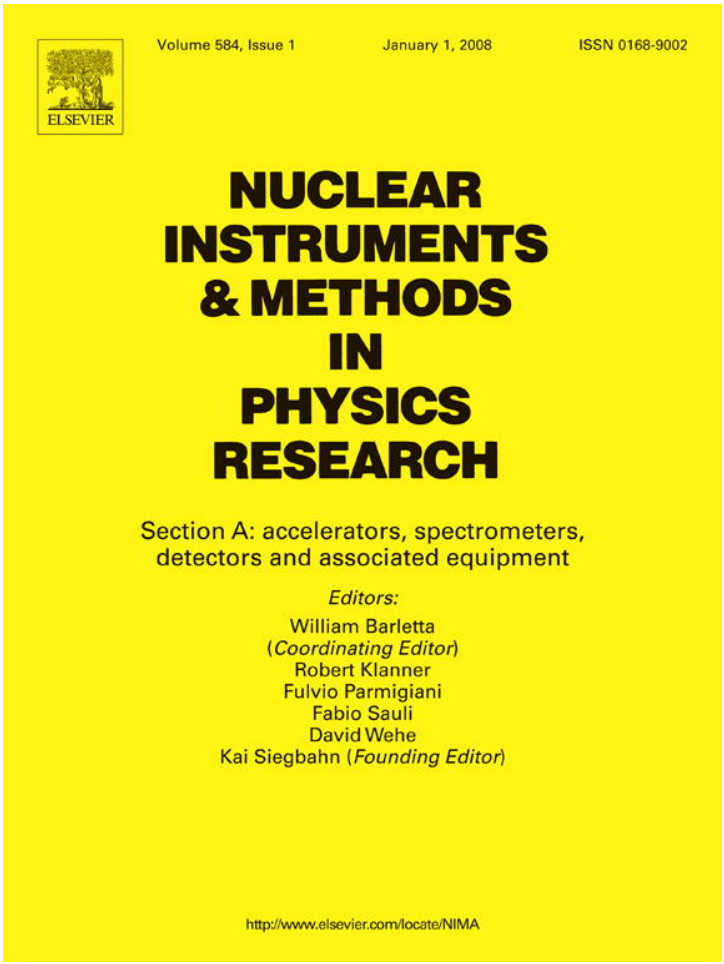

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Pulse-shape discrimination with $PbWO_4$ crystal scintillators

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Abstract

Light output, α/β ratio, and pulse shape have been investigated at $-25\,^{\circ}C$ with $PbWO_4$ crystal scintillators undoped, and doped by F, Eu, Mo, Gd and S. The fast 0.01–0.06 μs and middle 0.1–0.5 μs components of scintillation decay were observed for all the samples. Slow components of scintillation signal with decay times 1–3 and 13–28 μs with total intensity up to ≈ 50% have been recognized for several samples doped by Molybdenum. We found some indications of a pulse-shape discrimination between α particles and γ quanta with $PbWO_4$ (Mo doped) crystal scintillators.

PACS: 29.40.Mc

Keywords: Scintillation detector; $PbWO_4$ crystals; Doping; Pulse-shape discrimination

1. Introduction

Lead tungstate crystals ($PbWO_4$) are discussed in Ref. [1] as promising material for a high sensitivity experiment to search for double beta decay of ^{116}Cd with the help of cadmium tungstate crystal scintillators ($CdWO_4$). $PbWO_4$ crystals can be used as light-guides and high efficiency active shield for low-background $CdWO_4$ scintillation detectors. Using of radiopure $PbWO_4$ scintillators could allow to build a 2β experiment to search for 0ν2β decay of ^{116}Cd at the level of sensitivity $T^{0\nu2\beta}_{1/2} \sim 10^{26}$ yr, which corresponds to the Majorana neutrino mass ~0.1–0.05 eV.

As it has been demonstrated (see, for instance Refs. [2–4]), a pulse-shape discrimination ability of the scintillation detector is important to interpret and to reject background caused by internal contamination of ^{232}Th and ^{238}U daughters. However, pulse-shape discrimination technique can be also useful to suppress background in a detector from internal contamination in surrounding scintillator (for instance, γ background in the $CdWO_4$ detector from U/Th contamination in the $PbWO_4$ surrounding crystals [1]). Pulse-shape discrimination in $PbWO_4$ scintillators can be useful to suppress background γ events in the $CdWO_4$ detector from ^{208}Tl decays in $PbWO_4$ by off line tagging α events from the preceding ^{212}Bi decay. Similarly the γ events from ^{214}Bi β decay can be rejected by tagging subsequent ^{214}Po α events in $PbWO_4$ scintillators.

The aim of the present study was to check a possibility of pulse-shape discrimination with $PbWO_4$ crystal scintillators. In addition, the influence of fluorine doping on light yield of $PbWO_4$ scintillators reported in Ref. [5] has been checked, and response of crystals with different dopants to α particles has been investigated.

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Table 1
Parameters of the $PbWO_4$ samples

Sample	Dopants	Concentration in the melt (ppm)	Concentration of Mo, Eu, and Gd determined by SIMS (ppm)
20/01	–	–	35 (Mo), ⩽3 (Eu), ⩽1 (Gd)
6/03	F	400	35 (Mo), ⩽3 (Eu), ⩽1 (Gd)
8/03	F	4000	10 (Mo), ⩽3 (Eu), ⩽1 (Gd)
9/03	F, Gd	2000, 1000	35 (Mo), ⩽3 (Eu), 100 (Gd)
7/03	F, Eu	400, 100	35 (Mo), 25 (Eu), ⩽1 (Gd)
13/03	F, Eu	2000, 20	40 (Mo), 33 (Eu), ⩽1 (Gd)
10/03	F, Gd, Mo	1700, 90, 15 800	12 000 (Mo), ⩽3 (Eu), 40 (Gd)
11/03	F, Gd, Mo	1000, 50, 30 000	26 000 (Mo), ⩽3 (Eu), 15 (Gd)
12/03	F, Gd, Mo	870, 40, 78 800	55 000 (Mo), ⩽3 (Eu), 27 (Gd)
14/03	F, Eu, Mo	1000, 6, 30 000	18 000 (Mo), 5 (Eu), ⩽1 (Gd)
15/03	F, Eu, Mo, S	820, 5, 24 500, 1000	15 000 (Mo), 10 (Eu), ⩽1 (Gd)

2. Measurements and results

2.1. Samples

$PbWO_4$ single crystals were grown using Czochralski technique in an inert (Argon) atmosphere. Dopants (RE_{3+} ions) were added to the raw material ($PbO + WO_3$) in the form of RE_2O_3 oxides. Fluorine, molybdenum and sulfur were in the melt as a PbF_2, MoO_3 and $PbSO_4$, correspondingly. Samples for measurements were cut from crystal boules and polished in the form of plates with a diameter of 10–15 mm and a thickness of ≈ 5 mm. The $PbWO_4$ crystals used in the present research are listed in Table 1. All the crystals were colorless and transparent except one (6/03), which was slightly yellow colored. Some of the samples have visible core-like defects. Concentration of Mo, Eu, Gd dopant elements was checked with the help of Secondary Ion Mass Spectroscopy (SIMS, Cameca, IMS-4F). The device was set to the mode of operation when positive charged ions were detected, therefore we were not able to estimate concentrations of F and S in the samples. The results of the mass-spectrometry (provided in Table 1) qualitatively confirm the concentration of the dopant elements known from the growth conditions.

2.2. Relative light output

The samples of $PbWO_4$ scintillators were wrapped by PTFE reflector tape and optically connected to the PMT EMI9256KB by Dow Corning Q2-3067 optical couplant. The detector was placed into temperature controlled chamber, where the temperature of the detector was stabilized and measured with an accuracy of $\pm 1\,^{\circ}C$. The shaping time of the ORTEC (Model 572) amplifier was set to 0.5 μs. The relative light output of the $PbWO_4$ detectors was measured at $-25\,^{\circ}C$ with ^{60}Co and ^{137}Cs γ sources.

Fig. 1(a) shows the energy spectra of ^{57}Co and ^{137}Cs γ sources measured with the crystal $PbWO_4$ doped by F, Gd and Mo (10/03). An energy resolution FWHM = 23% was obtained for 662 keV γ line of ^{137}Cs.

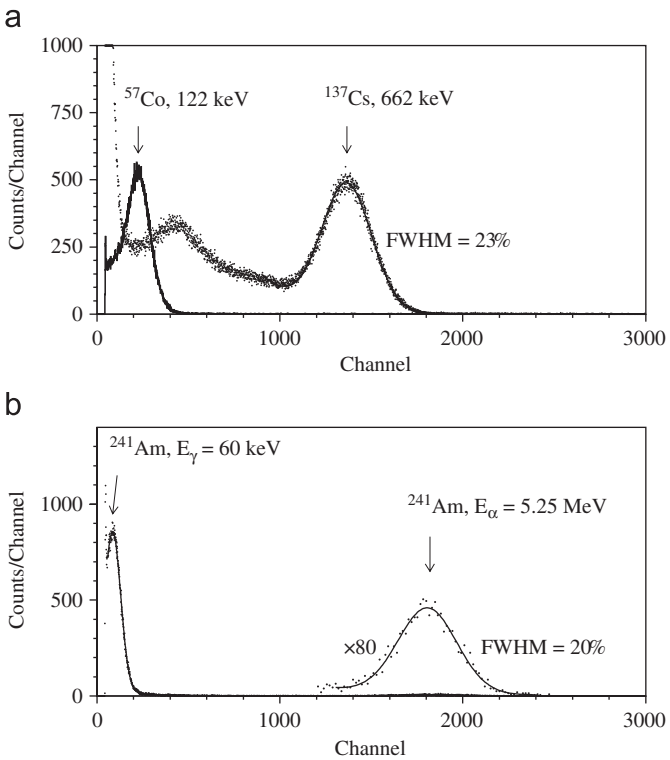

Fig. 1. Energy spectra measured by the $PbWO_4$(F,Gd,Mo) scintillation crystal (10/03) with (a) ^{57}Co and ^{137}Cs sources and (b) ^{241}Am source at $-25\,^{\circ}C$.

The spectrum measured with ^{241}Am α source is shown in Fig. 1(b). It should be stressed that even the 60 keV γ peak of ^{241}Am is still resolved from the PMT noise. The energy resolution for the 60 keV γ peak is FWHM ≈ 87%.

The samples of $PbWO_4$ scintillators were prepared from crystal boules of different diameter (10–15 mm) and have values of height-to-diameter ratio (h/D) in the range 0.3–0.5. Relative light output of scintillators with high index of refraction (the index of refraction of $PbWO_4$ crystals is 2.2) depends on the h/D ratio. To take into account such an effect, we have carried out measurements with $CdWO_4$ crystal scintillators (index of refraction in the

range 2.1–2.3) with the h/D ratio in the range 0.06–0.9. A $CdWO_4$ crystal $\oslash 15 \times 13.5$ mm (therefore, an initial value was $h/D = 0.9$) was used for the measurements. To obtain the samples with lower h/D ratio, the crystal was cut on the cleavage plane. The $CdWO_4$ scintillators were covered by teflon reflector and optically coupled to Philips XP2412 photomultiplier. The measurements were carried out with a ^{207}Bi gamma source. The results of measurements are shown in Fig. 2. The data were fitted by the polynomial function $f(h/D) = p_0 + p_1 \cdot (h/D) + p_2/(h/D)$. Then the values of relative light output measured with $PbWO_4$ scintillators were corrected by using the experimentally determined coefficients. The relative light output measured with the $PbWO_4$ samples after correction on the height-to-diameter ratio (corrections do not exceed 10%) are presented in Table 2. It should be stressed that emission

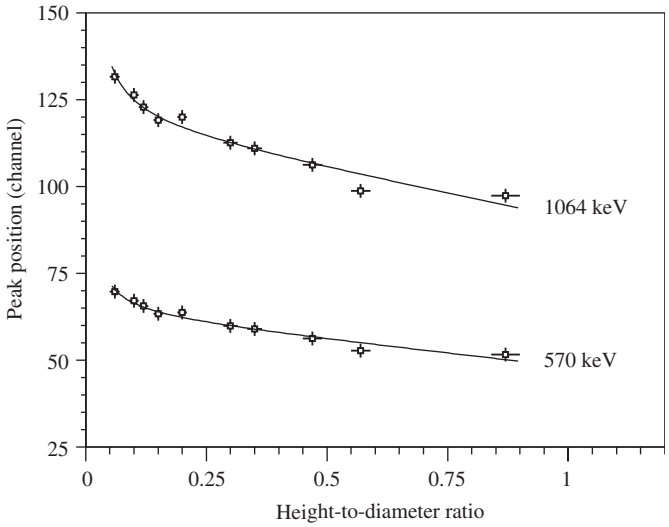

Fig. 2. Dependence of relative pulse amplitude on sample height-to-diameter ratio measured with $CdWO_4$ crystal scintillator. The solid line represents the fits of the data by polynomial function.

Table 2
Relative light output, and α/β ratio measured both with an external ^{241}Am α source and by using the α peak of ^{210}Po from internal contamination of the $PbWO_4$ samples

Sample, dopants	Relative light output	α/β ratio	
		External α source	^{210}Po
20/01 (undoped)	1.00	0.24(3)	0.22(3)
6/03 (F)	0.94	0.21(3)	0.21(3)
8/03 (F)	0.98	0.22(3)	0.20(3)
9/03 (F,Gd)	0.81	0.26(4)	0.25(4)
7/03 (F,Eu)	0.73	0.19(4)	–
13/03 (F,Eu)	0.92	0.22(3)	0.21(3)
10/03 (F,Gd,Mo)	2.98	0.32(3)	0.29(3)
11/03 (F,Gd,Mo)	1.92	0.23(3)	0.22(3)
12/03 (F,Gd,Mo)	1.35	0.21(3)	0.22(3)
14/03 (F,Eu,Mo)	1.83	0.23(3)	0.22(3)
15/03 (F,Eu,Mo,S)	1.73	0.21(3)	0.22(3)

spectra and light attenuation length of $CdWO_4$ and $PbWO_4$ crystals are different. Therefore, the correction factors extracted from the measurements with the $CdWO_4$ samples can be considered as the first order approximation for $PbWO_4$ scintillators. Besides, one should take into account that the relative light output depends on scintillation emission spectra and decay kinetics of $PbWO_4$ scintillators [6]. By using PMT with other spectral sensitivity and/or spectroscopy amplifier with other shaping time one could obtain slightly different results.

We do not observe an increasing of light output for $PbWO_4$ scintillators grown by Czochralski method and doped by fluorine, as reported in Ref. [5]. This result is in agreement with Ref. [7]. Recently a factor of 2.5 increase of the light yield was reported for F doped $PbWO_4$ crystals grown by Bridgman method [8]. In opinion of authors of Ref. [8] "F-ion does not enter substantially in Czochralski-grown PWO, and any improvement has not been observed". Therefore, additional investigations are necessary to understand how the growth methods affect the properties of $PbWO_4$ crystals. It should be stressed that only initial concentrations of the dopants in the raw materials were known in all cases. A careful quantitative control of dopant elements, as well as impurity concentrations in the crystals are required for such investigations.

2.3. α/β ratio

The α/β ratio was measured with the help of the collimated α particles of an ^{241}Am source. As it was checked by surface-barrier silicon detector, the energy of α particles was reduced to about 5.25 MeV going through 2 mm of air in the collimator. The energy scale was calibrated with the help of the 570 and 1064 keV γ lines of a ^{207}Bi source. The results are presented in Table 2. The α/β ratio measured with the external α source are in the range of 0.20–0.32 which is an agreement with result reported in Ref. [1]. The errors of the α/β ratio are mainly due to uncertainties in the energy calibration of the $PbWO_4$ detectors due to the poor energy resolution of the γ peaks.

In addition, the α peak of ^{210}Po (from internal contamination of crystals by ^{210}Pb) was used to measure the α/β ratio. The energy spectrum of $PbWO_4$ scintillator (sample 10/03) measured during 2600 s is presented in Fig. 3. A clear peak in the spectrum can be attributed to α decay of ^{210}Po (the energy of α particles is 5.31 MeV) from internal contamination of the crystal by ^{210}Pb. The α/β ratio is 0.29 ± 0.03, the activity of ^{210}Po in the crystal is 24 ± 2 Bq/kg.

As one can see in Table 2, the values obtained with an external α source and by using the peak of internal ^{210}Po are in agreement. Some difference in the values of the α/β ratio between the studied $PbWO_4$ samples was observed. However, we cannot interpret the difference as own properties of the samples. This is because the α/β ratio is not only a property of the crystal material, but also a certain characteristics of the scintillation detector

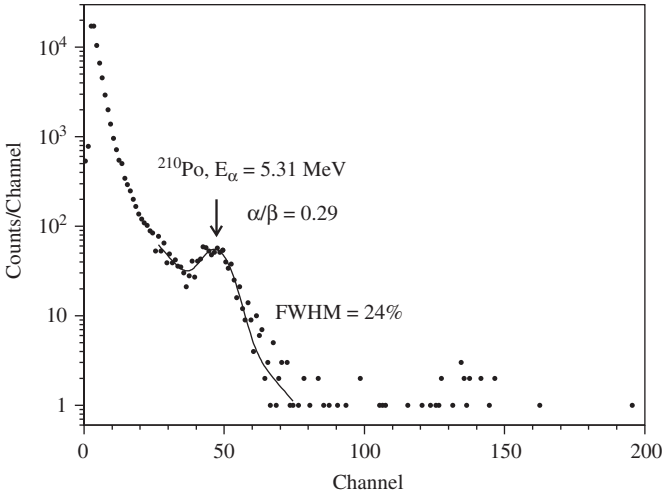

Fig. 3. Energy spectrum of $PbWO_4$ scintillation crystal (10/03) measured during 2600 s. The clear peak in the spectrum at the energy ≈ 1.54 MeV (in γ scale) can be attributed to α decay of ^{210}Po from internal contamination of the crystal by ^{210}Pb. The α/β ratio is 0.29.

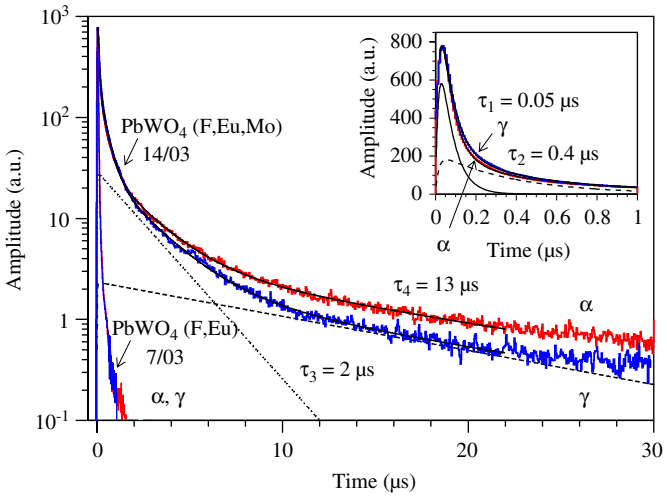

Fig. 4. Decay of scintillation of the $PbWO_4$ crystal (14/03, doped by F, Eu, Mo) for γ quanta and α particles and their fit by four components. Pulse shapes of $PbWO_4$ scintillator doped by F and Eu (7/03) are shown for comparison. (Inset) The first part of the $PbWO_4$ (14/03) α and γ pulses.

depending on the shape, sizes, surface treatment, transparency of a crystal, etc. To check a possible effect of dopant elements on the α/β ratio in $PbWO_4$ scintillators, one has to provide the same shape, sizes, transparency (without visible defects) and surface quality of samples.

2.4. Pulse-shape discrimination for γ quanta and α particles

2.4.1. Shapes of scintillation light pulses

The shapes of scintillation light pulses in the $PbWO_4$ crystals were studied for α particles from ^{241}Am source and for γ quanta from ^{137}Cs and ^{60}Co sources with the help of the 125 MSample/s 12 bit digitizer described in Ref. [9]. The shapes of scintillation pulses in the sample 13/03, and in the samples doped by molybdenum were also measured in a time interval ≈ 80 μs with the help of the 20 MSample/s 12 bit digitizer [10]. To determine shapes of scintillation pulses for the sample 13/03, and for the samples with molybdenum, the data obtained with both the 20 and 125 MSample/s digitizers were used. The pulse shapes were constructed in the time interval 0–1.5 μs from the 125 MSample/s data, and in the interval 1.5–50 μs from the data measured by the 20 MSample/s device. The pulse shapes for γ quanta and α particles measured with the $PbWO_4$ crystal (7/03) doped by F and Eu, and (14/03) doped by F, Eu, Mo are depicted in Fig. 4. The pulse shape can be fitted by a sum of exponential functions:

$$f(t) = \sum A_i(e^{-t/\tau_i} - e^{-t/\tau_0})/(\tau_i - \tau_0), \quad t > 0$$

where A_i are intensities, and τ_i are decay constants for different light emission components, τ_0 is the integration constant of electronics (≈ 0.02 μs). The values of A_i and τ_i obtained by fitting the average of a few thousand individual α and γ pulses are presented in Table 3.

The 0.01–0.06 μs and 0.1–0.5 μs decay components are in agreement with the results obtained in Refs. [11–18]. The slow 1–3 μs decay of $PbWO_4$ scintillation was also observed in Refs. [6,12,13,19], while 13–28 μs decay components were never reported for $PbWO_4$ scintillators. We were not able to measure the nanosecond (≈ 1–9 ns) decay component, because of a comparatively large value of integration constant of the electronics ≈ 0.02 μs used in the present study.

2.4.2. Pulse-shape discrimination by optimal filter method

A small difference in light pulse shapes of $PbWO_4$ crystal scintillators could allow to discriminate γ(β) events from α particles. We applied for this purpose two approaches: the optimal filter technique proposed in Ref. [20] and developed in Ref. [10] for $CdWO_4$ crystal scintillator, and the mean time method.

To obtain a numerical parameter for the $PbWO_4$ signal, the so-called shape indicator (SI), the following formula was applied for each pulse:

$$SI = \sum f(t_k)P(t_k) \Big/ \sum f(t_k)$$

where the sum is over time channels k, starting from the origin of the pulse up to certain time, $f(t_k)$ is the digitized amplitude (at the time t_k) of a given signal. The weight function $P(t)$ is defined as: $P(t) = \{f_\alpha(t) - f_\gamma(t)\}/\{f_\alpha(t) + f_\gamma(t)\}$, where $f_\alpha(t)$ and $f_\gamma(t)$ are the reference pulse shapes for α particles and γ quanta. Reasonable discrimination between α particles and γ quanta was achieved with Molybdenum doped $PbWO_4$ crystals as one can see in Fig. 5 where the SI distributions measured by the $PbWO_4$(F,Gd,Mo) scintillation crystal (10/03) with α particles (≈ 5 MeV) and γ quanta (≈ 1.3 MeV) are shown. The small tail in the SI distribution for γ at SI ≈ 50 can be

Table 3
The time properties of $PbWO_4$ crystal scintillators

Sample, dopants	Type of irradiation	Decay constants (μs) and relative intensities				FOM	
		τ_1 (A_1)	τ_2 (A_2)	τ_3 (A_3)	τ_4 (A_4)	OF	MT
20/01	γ	0.016 (72%)	0.24 (17%)	2.6 (11%)	–	0.05	0.02
(undoped)	α	0.016 (74%)	0.22 (16%)	1.9 (11%)	–		
6/03	γ	0.012 (77%)	0.29 (12%)	1.1 (11%)	–	0.08	0.06
(F)	α	0.013 (78%)	0.24 (11%)	1.0 (11%)	–		
9/03	γ	0.026 (86%)	0.13 (14%)	1–5 (<0.5%)	–	0.03	0.01
(F,Gd)	α	0.031 (84%)	0.15 (16%)	1–5 (<0.5%)	–		
7/03	γ	0.014 (95%)	0.24 (5%)	1–5 (<0.5%)	–	0.04	0.01
(F,Eu)	α	0.016 (94%)	0.19 (6%)	1–5 (<0.5%)	–		
13/03	γ	0.026 (79%)	0.5 (13%)	2.0 (7%)	≈ 17 (1%)	0.48	0.35
(F,Eu)	α	0.028 (84%)	0.4 (10%)	1.8 (5%)	≈ 18 (1%)		
10/03	γ	0.064 (42%)	0.30 (46%)	2.3 (8%)	20 (4%)	2.4	1.6
(F,Gd,Mo)	α	0.061 (33%)	0.37 (40%)	2.6 (14%)	28 (12%)		
11/03	γ	0.059 (25%)	0.40 (32%)	2.5 (24%)	16 (19%)	1.4	1.2
(F,Gd,Mo)	α	0.047 (23%)	0.36 (26%)	2.1 (25%)	15 (26%)		
12/03	γ	0.058 (25%)	0.38 (27%)	2.4 (24%)	17 (24%)	1.8	1.4
(F,Gd,Mo)	α	0.048 (22%)	0.35 (23%)	2.1 (23%)	16 (32%)		
14/03	γ	0.062 (25%)	0.38 (35%)	2.1 (27%)	13 (13%)	2.3	1.5
(F,Eu,Mo)	α	0.054 (24%)	0.35 (28%)	2.0 (27%)	13 (21%)		
15/03	γ	0.064 (26%)	0.38 (34%)	2.1 (27%)	14 (12%)	1.6	1.4
(F,Eu,Mo,S)	α	0.054 (25%)	0.34 (28%)	1.7 (27%)	13 (20%)		

The decay constants and their intensities (in percentage of the total intensity) for γ quanta and α particles are denoted as τ_i and A_i, respectively. The factor of merit for pulse-shape discrimination (FOM, see text) characterizes the efficiency of pulse-shape discrimination with the help of the optimal filter (OF), and mean time (MT) methods.

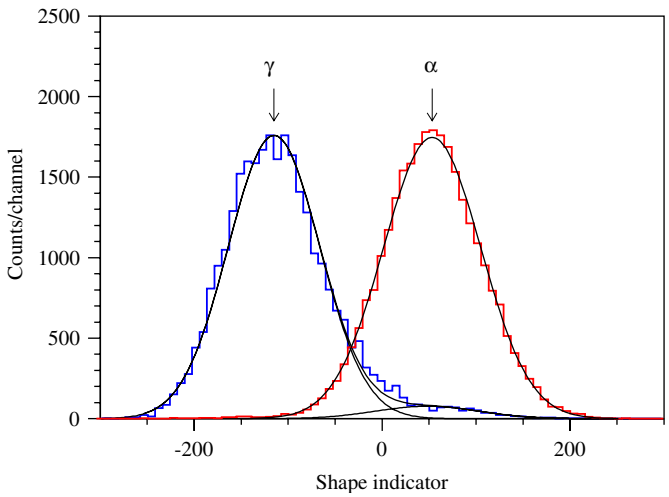

Fig. 5. Distributions of the shape indicator (SI, see text) for pulses produced by γ quanta and α particles in $PbWO_4$(F, Gd, Mo) scintillation crystal (10/03). The small tail in the shape indicator distribution for γ at SI ≈ 50 can be explained by background α events from ^{210}Po inside the crystal.

explained by background α events from ^{210}Po inside the crystal (see Section 2.3 and Fig. 3).

As a measure of discrimination ability (factor of merit, FOM), the following formula can be used:

$$\mathrm{FOM} = |\mathrm{SI}_\alpha - \mathrm{SI}_\gamma| \Big/ \sqrt{\sigma_\alpha^2 + \sigma_\gamma^2}$$

where SI_α and SI_γ are mean SI values for α particles and γ quanta distributions (which are well described by Gaussian functions), σ_α and σ_γ the corresponding standard deviations. For the distributions presented in Fig. 5, the factor of merit is FOM = 2.4.

2.4.3. Mean time method

The data were processed also by using the mean time method. The following formula was applied to calculate the parameter $\langle t \rangle$ (mean time) for each pulse:

$$\langle t \rangle = \sum f(t_k) t_k \Big/ \sum f(t_k)$$

where the sum is over time channels k, starting from the origin of pulse and up to a certain time. The distributions of parameters $\langle t \rangle$ are well described by Gaussian functions. Therefore the factor of merit FOM can be used to characterize efficiency of pulse-shape discrimination. As one can see from Table 3, the optimal filter method provides slightly better pulse-shape discrimination. At the same time the mean time method is easy to apply because it does not need the construction of a weight function.

3. Conclusions

The light output, α/β ratio, and pulse shape have been investigated at −25 °C with $PbWO_4$ crystal scintillators undoped, and doped by F, Eu, Mo, Gd and S. Doping of $PbWO_4$ crystals by molybdenum improves light output by

approximately a factor of 1.5–3. This result is in agreement with data published before [21,22]. The relative light output of europium doped $PbWO_4$ crystals is on the level of 0.7–0.9 relatively to undoped sample. We do not observe an increase of light output by fluorine doping as reported in Refs. [5,8]. This fact can be explained by different methods used to grow $PbWO_4$(F) crystals: Bridgman in Refs. [5,8], and Czochralski in the present study and in the work [7] where the effect was not observed too.

Values of the α/β ratio measured with different $PbWO_4$ samples vary in the range of 0.20–0.32. However, such a difference can be explained by unequal quality of the samples that have been used in the measurements. To study the possible effect of doping on the α/β ratio one should prepare the samples with the same sizes, surface quality, and without visible defects.

Fast 0.01–0.06 μs and middle 0.1–0.5 μs components of scintillation decay were observed for all the samples. Slow components of scintillation signal with the decay times 1–3, and 13–28 μs with the total intensity at the level of $\approx$ 45–50% have been measured for samples doped by Molybdenum. The undoped crystal as well as crystals without molybdenum have shown a lower intensity of the slow components at the level of $\leqslant$0.5–10%.

We found some indications of a pulse-shape discrimination between α particles and γ quanta by applying the mean time and optimal filter methods. The best discrimination was achieved with $PbWO_4$ crystals doped by molybdenum.

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