Scintillation pulse shape discrimination with $CaWO_4$, $ZnWO_4$, and $CdWO_4$ crystal

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The time properties of scintillation pulse form for γ rays and α particles in $CaWO_4$, $ZnWO_4$, and $CdWO_4$ crystal have been investigated. A method for discrimination of various particle types using the pulse shape distinctions has been developed. A complete signal separation for γ quanta and α particles has been attained. The pulse shape dependences on the α irradiation energy (for all the crystals) and direction in relation to the main crystallographic axes (for $ZnWO_4$ and $CdWO_4$ scintillators) have been observed and studied. The temperature dependence of the average decay time of $CdWO_4$ for γ rays and α particles has been measured in temperature interval 0–30°C.

Временные свойства сцинтилляционных импульсов для γ квантов и α частиц были исследованы для кристаллов $CaWO_4$, $ZnWO_4$, $CdWO_4$. Благодаря зависимости формы импульса от типа ионизирующего излучения разработан метод идентификации сигналов, основанный на методе оптимального цифрового фильтра. Получено полное разделение α-частиц и γ-квантов для данных кристаллов. Измерены зависимости формы сцинтилляционного импульса от энергии и от направления облучения относительно главных кристаллографических осей. Для кристалла $CdWO_4$ получена зависимость среднего времени высвечивания для γ-квантов и α-частиц от температуры в интервале 0–30°C.

The pulse shape discrimination ability is an important characteristic of low counting experiments aimed to search for extraordinary rare effects, such as double beta (2β) decay [1], dark matter particles [2], rare β and α decays. For instance, in the experiment [3], an extremely low background level of 0.04 counts/(yr·keV·kg) was attained with cadmium tungstate ($CdWO_4$) crystal scintillator due to the pulse shape discrimination. The half-life limit on 0ν2β decay of ^{116}Cd ($T_{1/2} \geq 1.7\cdot10^{23}$ yr at 90 % C.L.) was set, which results in one of the strongest restrictions on the effective Majorana neutrino mass: $\langle m_\nu \rangle \leq 1.7$ eV. With the same crystals, using also the pulse shape discrimination, the first indication for the α decay of ^{180}W with the half-life $T_{1/2} = 1.1\cdot10^{18}$ yr has been observed [4]. The aim of this work was to study the pulse shape discrimination ability of $CaWO_4$, $ZnWO_4$ and $CdWO_4$ crystal scintillators.

The main properties of the studied scintillators are presented in Table 1. The crystals were grown by the Czochralski method. All these crystals are chemically resistant and non-hygroscopic. The $CaWO_4$ crystal possesses better mechanical properties than $ZnWO_4$ and $CdWO_4$ ones. The crystals were wrapped by PTFE reflecting tape and optically coupled to a PMT (Philips XP2412 with bialkali photocathodes and EMI D724KFLB with RbCs one). A data acquisition system records the pulse shape of the scintillation signals using a transient digitizer based on a 12 bit ADC (AD9022) with the sampling frequency of 20 MHz. The pulses of events in the selected energy interval were recorded in 2048 time channels at 50 ns channel width.

Pulse shapes for α particles and γ quanta (^{232}Th) in a wide energy range were investigated. A collimated ^{241}Am source with thin Mylar films (0.653 mg/cm^2) and air layer as absorbers was used. Alpha particles in the energy range of 0.5 to 5.3 MeV were obtained. The energy of α particles was determined using a surface-barrier detector. The crystals were irradiated with α particles in the directions perpendicular to (010), (001), and (100) crystal planes to study the dependence of pulse shape on the irradiation direction. In addition, α events from the decay of nuclides from the ^{232}Th, ^{235}U, and ^{238}U chains present as traces in the $CdWO_4$ crystal were used to extend the energy range up to about 8 MeV. These events were selected from data of low background measurements using the time-amplitude analysis [5, 6].

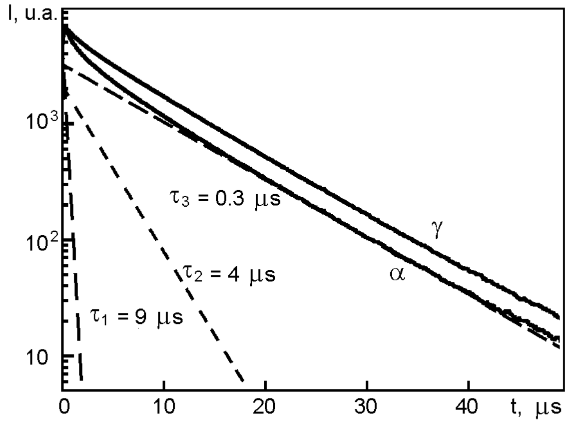

Fig. 1. Shapes of scintillation pulses (I, intensity in arbitrary units) in $CaWO_4$ crystal for γ rays and α particles (average of 4000 pulses) and their fits by three exponential functions with decay constants 0.3 μs, 4 μs, and 9 μs.

The time characteristics of $CaWO_4$ (size $40\times34\times23$ mm^3), $ZnWO_4$ ($\varnothing14\times7$ mm^2), $CdWO_4$ ($\varnothing32\times19$ mm^2) scintillators were studied as described in [4, 7, 8]. To describe the scintillation signals analytically, the pulse shape resulting from the averaging of a large number of individual events was fitted by function (see Fig. 1):

$$f(t) = \sum_i \frac{A_i}{(\tau_i - \tau_0)} \cdot \left(e^{-\frac{t}{\tau_i}} - e^{-\frac{t}{\tau_0}}\right), \quad t > 0,$$

where A_i are the relative intensities (as percentage of the total intensity); τ_i, the decay constants for various light emission components; τ_0, the integration constant of electronics (≈ 0.2 μs). The fitting exhibited three decay components with different in-

Table 1. Properties of $CaWO_4$, $ZnWO_4$ and $CdWO_4$ scintillators. The effective average decay time and relative photoelectron yield were measured at room temperature for γ quanta

	$CaWO_4$	$ZnWO_4$	$CdWO_4$
Density (g/cm^3)	6.1	7.8	8.0
Melting point (°C)	1570–1650	1200	1325
Structural type	Sheelite	Wolframite	Wolframite
Cleavage plane	Weak (101)	Marked (010)	Marked (010)
Hardness (Mohs)	4.5–5	4–4.5	4–4.5
Emission maximum (nm)	420–425	480	480
Refractive index	1.94	2.1–2.2	2.2–2.3
Effective average decay time (μs)	8	24	13
Photoelectron yield [% of NaI(Tl)]	18 %	13 %	20 %

Table 2. Time properties of $CaWO_4$, $ZnWO_4$ and $CdWO_4$ scintillators (at room temperature). The decay constants and their intensities (in percentage of the total intensity) are denoted as τ_i and A_i, respectively

Crystal	Type of irradiation	Decay constants (τ_i), μs		
		τ_1 (A_1)	τ_2 ($A_{2\,nothingE\#*32B}$)	τ_3 (A_3)
$CaWO_4$	γ rays	0.3 (3 %)	4.4 (15 %)	9.0 (82 %)
	α particles	0.3 (6 %)	3.2 (18 %)	8.8 (76 %)
$ZnWO_4$	γ rays	0.7 (2 %)	7.5 (9 %)	25.9 (89 %)
	α particles	0.7 (4 %)	5.6 (16 %)	24.8 (80 %)
$CdWO_4$	γ rays	–	2.1 (5.7 %)	13.6 (94.3 %)
	α particles	1.0 (5.7 %)	3.3 (13.4 %)	12.7 (80.9 %)

tensities for γ quanta and α particles (see Table 2).

The difference in the pulse-shapes allows to discriminate γ quanta from α particles. To that purpose, we applied the optimal filter method proposed in [9] and developed in [10]. To obtain the numerical characteristic of scintillation signal, so-called shape indicator (SI), the following formula was applied for each pulse:

$$SI = \frac{\sum f(t_k) \cdot P(t_k)}{\sum f(t_k)},$$

where the summation is done over time channels k, starting from the pulse origin and up to 75 μs for $CaWO_4$ and $CdWO_4$ crystals, and up to 90 μs for $ZnWO_4$, $f(t_k)$ is the digitized amplitude (at the time t_k) of a given signal. The weight function $P(t)$ is defined as:

$$P(t) = \frac{f_\alpha(t) - f_\gamma(t)}{f_\alpha(t) + f_\gamma(t)},$$

where $f_\alpha(t)$ and $f_\gamma(t)$ are the reference pulse shapes for α particles and γ rays, respectively. A clear discrimination between α particles and γ quanta was achieved using this approach, as is seen in Fig. 2, where the shape indicator distributions measured for the $CaWO_4$ scintillation crystal with α particles ($E_\alpha \approx 5.3$ MeV) and γ quanta (≈ 1 MeV) are shown. The SI distributions are well described by Gaussian functions, the standard deviations σ_α and σ_γ being energy-dependent [4, 10]. As a measure of discrimination ability, the following quantity (the discrimination ability parameter) can be used:

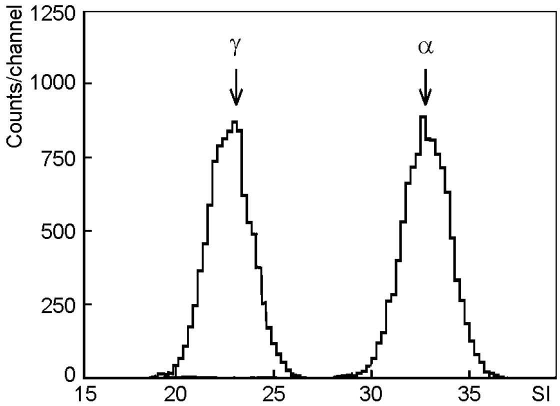

Fig. 2. The shape indicator (SI) distributions measured for $CaWO_4$ detector with γ quanta (≈ 1.2 MeV) and α particles (E_α = 5.3 MeV).

$$M = \frac{|SI_\alpha - SI_\gamma|}{\sqrt{\sigma_\alpha^2 + \sigma_\gamma^2}},$$

where SI_α and SI_γ are the mean SI values of α particles and γ quanta distributions; σ_α and σ_γ, the corresponding standard deviations. For the distributions presented in Fig. 2, the parameter $M = 5.9$. For $ZnWO_4$ and $CdWO_4$ crystals, the M values 5.2 and 6.7, respectively, have been obtained. The discrimination ability becomes worse with decreasing of α particles and γ quanta energy.

The energy dependence of the SI for $CaWO_4$ was studied for α particles in the 1–5.3 MeV range (see Fig. 3). The shape indicator measured for $CaWO_4$ crystals with α particles does not depend on the α irradiation direction relative to the crystal planes. The energy dependence of the SI for $CdWO_4$ and $ZnWO_4$ was studied for α particles in the 0.5–8.0 MeV and the 2–5.3 MeV ranges, respectively. The dependence of SI

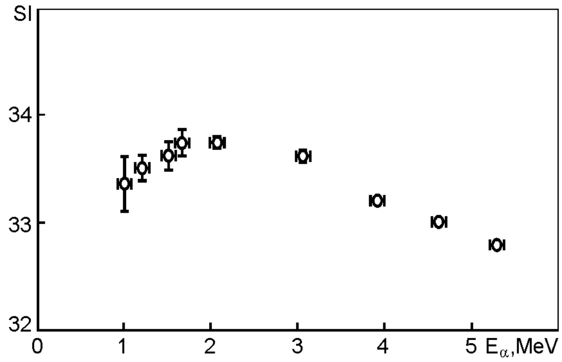

Fig. 3. The energy dependence of the shape indicator measured for $CaWO_4$ crystal scintillator for α particles in the 1–5.3 MeV energy range.

on the irradiation energy and direction was observed for α particles [4, 8]. For instance, the energy and direction dependences of SI for $ZnWO_4$ crystal scintillator are shown in Fig. 4. No energy dependence of the SI for γ quanta in the range from 0.1 to 2.6 MeV was observed for $CaWO_4$, $ZnWO_4$, and $CdWO_4$ crystals.

The measured temperature dependence of average decay time for $CdWO_4$ signals in the 0–30°C range is shown in Fig. 5. The behavior of experimental points agrees with that from [11]. It should be noted that the average time decreases as the temperature rises. The discrimination ability parameter M shows a slight increase with temperature.

The three time components $\tau_i \approx 0.7$, ≈ 7 and ≈ 25 μs were observed for $ZnWO_4$ scintillation signals. The value for the slow decay component was in agreement with the result (25 μs) obtained in [12], while 0.7 and 7 μs decay time constants were identified for the first time. We were not able to recognize the 100 ns decay component reported in [12] because of a rather slow (20 MHz) transient digitizer used in measurements. For $CaWO_4$ crystals, the best fit was achieved with three decay components (0.3, 4, and 9 μs).

The energy dependence of the shape indicator (SI) for α particles can be explained by the dependence of the ionization density on the energy.

The $CaWO_4$ [13, 14], $ZnWO_4$ [8], and $CdWO_4$ [15] crystal scintillators were proposed as bolometric detectors for investigation of rare nuclear decays and dark matter search. The dependence of pulse shape on direction of high ionizing α particles (supposing that such a behavior would remain for nuclear recoils) observed with $ZnWO_4$

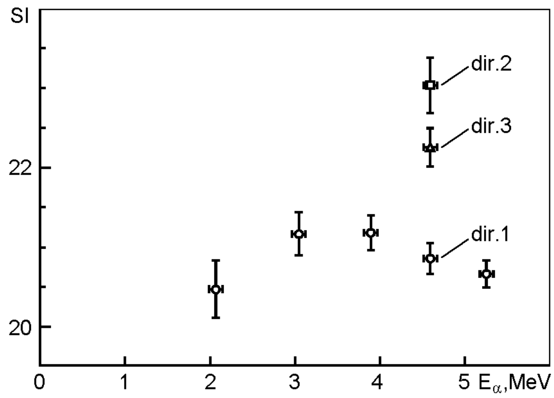

Fig. 4. The energy and direction dependence of the shape indicator measured for $ZnWO_4$ crystal scintillator for α particles in the energy range 2.0–5.3 MeV. The $ZnWO_4$ crystal was irradiated perpendicularly to the crystal planes: (010) — direction 1, (001) — direction 2, and (100) — direction 3.

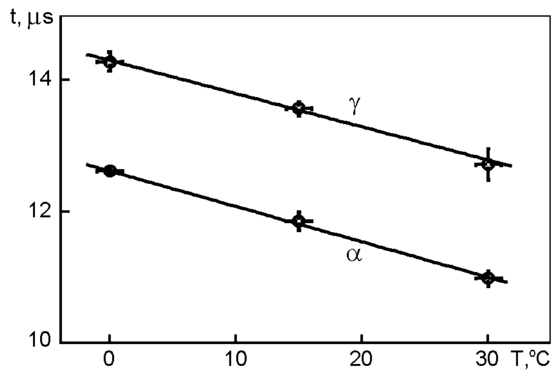

Fig. 5. The temperature dependence of the average decay time for $CdWO_4$ crystal scintillator measured in temperature range 0–30°C for γ quanta and α particles.

scintillator could be used to detect a diurnal asymmetry in observation of dark matter particles [2, 8].

To conclude, the pulse shape of $CaWO_4$, $ZnWO_4$ and $CdWO_4$ scintillation signals has been investigated and a good pulse shape discrimination for γ and α events has been attained. Dependence of the scintillation pulse shape on the energy of α particles have been observed. Besides, $ZnWO_4$ and $CdWO_4$ scintillators show a dependence of the pulse shape on the α irradiation direction relatively to the main crystal planes. The temperature dependence of the time properties and the discrimination ability parameter have been measured for the $CdWO_4$ crystal scintillators.

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Ідентифікація форми сцинтиляційних імпульсів для кристалів $CaWO_4$, $ZnWO_4$ та $CdWO_4$

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Часові характеристики сцинтиляційного спалаху для кристалів $CaWO_4$, $ZnWO_4$, $CdWO_4$ досліджено для γ-квантів та α-частинок. Розроблено метод дискримінації частинок різних типів, оснований на відмінності форми сцинтиляційного спалаху для частинок різних типів. Отримано повне розділення сигналів від α-частинок та γ-квантів. Досліджено залежності форми сигналу від енергії та напрямку опромінення відносно головних кристалографічних осей. Для кристала $CdWO_4$ отримано залежність середнього часу висвітлювання для γ-квантів та α-частинок від температури у діапазоні 0–30°C.