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ScienceDirect
Nuclear Instruments and Methods in Physics Research A 569 (2006) 743–753
NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH Section A
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Further study of $CdWO_4$ crystal scintillators as detectors for high sensitivity 2β experiments: Scintillation properties and pulse-shape discrimination

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Received 10 August 2006; received in revised form 31 August 2006; accepted 11 September 2006
Available online 20 October 2006

Abstract

Energy resolution, non-proportionality in the scintillation response, α/β ratio, pulse shape for γ rays and α particles were studied with $CdWO_4$ crystal scintillators. Some indication for a difference in the emission spectra for γ rays and α particles was observed. No dependence of $CdWO_4$ pulse shape on emission spectrum wavelengths under laser, α particles and γ ray excitation was observed. Dependence of scintillation pulse shape for γ quanta and α particles and pulse-shape discrimination ability on temperature was measured in the range of 0–24 °C.

PACS: 23.40.−s; 29.40.Mc

Keywords: Double beta decay; Scintillation detector; $CdWO_4$ crystals; Pulse-shape discrimination

1. Introduction

Observations of neutrino oscillations manifest the non-zero neutrino mass and provide important motivation for high sensitivity experiments on neutrinoless double beta (0ν2β) decay. However, this process still remains unobserved, and only half-life limits for 0ν2β mode were obtained (see, e.g., reviews [1]).[1] One of the most sensitive 2β experiments has been performed in the Solotvina Underground Laboratory [7] by the Kiev–Firenze collaboration with the help of enriched cadmium tungstate ($^{116}CdWO_4$) crystal scintillators [8,9]. The half-life limit on 0ν2β decay of ^{116}Cd was set as $T_{1/2} \geqslant 1.7 \times 10^{23}$ yr at 90% C.L., which corresponds to an upper bound on the effective Majorana neutrino mass $\langle m_\nu \rangle \leqslant 1.7$ eV [9]. This result is among the strongest world-wide restrictions on $\langle m_\nu \rangle$ (in addition to the bounds obtained in experiments with ^{76}Ge, ^{82}Se, ^{100}Mo, ^{130}Te, and ^{136}Xe).

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[1]An evidence for 0ν2β decay of ^{76}Ge has been claimed in Ref. [2]. However, it was criticized in Refs. [3–5]. Later the Heidelberg group has presented new data with improved statistics and after a reanalysis. A half-life $T_{1/2} \approx 1.2 \times 10^{25}$ yr has been reported [6], which corresponds to the effective Majorana neutrino mass $\langle m_\nu \rangle \approx 0.4$ eV.

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doi:10.1016/j.nima.2006.09.094

Two by-product results obtained in the course of the Solotvina experiment with $CdWO_4$ scintillators should also be mentioned: (i) the half-life ($T_{1/2} = 7.7 \pm 0.3 \times 10^{15}$ yr) and the spectrum shape of the fourth-forbidden β decay of ^{113}Cd were measured [10]; (ii) indication for the α decay of ^{180}W with the half-life $T_{1/2} = 1.1^{+0.9}_{-0.5} \times 10^{18}$ yr has been observed for the first time [11].

The Solotvina experiment demonstrates that $CdWO_4$ crystals possess several unique properties required for high sensitivity 2β decay experiments: low level of intrinsic radioactivity, good scintillation characteristics, and pulse-shape discrimination ability, which allow one to reduce background effectively. To enhance sensitivity to the neutrino mass to the level of $\langle m_\nu \rangle \sim 0.05$ eV, one has to increase the measuring time and the mass of enriched $^{116}CdWO_4$, improve the energy resolution and reduce background of the detector. As it was shown by Monte Carlo calculations, the required sensitivity could be achieved by using $^{116}CdWO_4$ crystals placed into a large volume of a high purity liquid. For instance, in the CAMEO project [12] it was proposed to place ≈ 100 kg of $^{116}CdWO_4$ crystals into the BOREXINO counting test facility. With 1000 kg of $^{116}CdWO_4$ crystals the neutrino mass limit can be pushed down to $\langle m_\nu \rangle \sim 0.02$ eV. An alternative solution should also be mentioned for a sensitive 2β decay experiment with $^{116}CdWO_4$ by using lead tungstate crystal scintillators as a high efficiency 4π active shield [13].

In addition, as it was demonstrated in Ref. [14], $CdWO_4$ crystals show good potential to develop thermal bolometers with energy resolution ≈ 5 keV in a wide energy interval. Furthermore, $CdWO_4$ can be used as a scintillating bolometer with registration of both light and heat signals [15]. Scintillating cryogenic detectors are highly promising to search for rare processes like dark matter and double beta decay thanks to their excellent energy resolution and particle discrimination ability.

Precise measurements of $CdWO_4$ properties are necessary for development of methods to simulate such detectors.

The purpose of our work was investigation of different $CdWO_4$ scintillation properties important for high sensitivity 2β experiment: energy resolution, non-proportionality in the scintillation response, α/β ratio, emission spectra and transparency, pulse shape for γ rays and α particles and their temperature dependence.

2. Measurements and results

The luminescence of $CdWO_4$ crystals was discovered about 60 years ago [16]. The different properties of $CdWO_4$ scintillators were investigated (see Refs. [17–32] and references therein). In 1960 Beard and Kelly have used a small natural $CdWO_4$ crystal to search for alpha activity of natural tungsten [33]. To our knowledge, it was the first low background experiment applying this detector.

The main characteristics of $CdWO_4$ scintillators are presented in Table 1. The material is non-hygroscopic and chemically resistant. All crystals used for measurements are listed in Table 2. All of them were grown by Czochralski method. The crystal CWO-1 was grown by the low-thermal-gradient Czochralski technique [34].

2.1. Scintillation properties

2.1.1. Energy resolution

In the present work, the energy resolution was measured with three $CdWO_4$ crystals: ⌀42 × 39 mm (CWO-1), ⌀40 × 30 mm (CWO-2), and 10 × 10 × 10 mm (CWO-3).

The CWO-1 crystal was ground at the side surface, the exit and top faces were polished. The crystal was wrapped by PTFE reflector tape and optically coupled to 3″ photomultiplier (PMT) Philips XP2412. The measurements were carried out with 16 μs shaping time to collect most of the charge from the anode of the PMT. The detector was irradiated by γ quanta of ^{60}Co, ^{137}Cs, ^{207}Bi, and ^{232}Th sources. The energy resolutions (full width at half maximum, FWHM) of 7.0% (^{137}Cs, 662 keV), 5.8% (^{207}Bi, 1064 keV), 5.0% (^{60}Co, 1333 keV), and 3.9% (^{232}Th, 2615 keV) were obtained (see Fig. 1).

The energy resolutions measured with the crystal CWO-2 in the same conditions are slightly worse. For instance, energy resolutions of 7.5%, 6.2%, and 4.6% were obtained

Table 1
Properties of $CdWO_4$ crystal scintillators

Density (g/cm^3)	7.9
Melting point (°C)	1271
Structural type	Wolframite
Cleavage plane	Marked (0 1 0)
Hardness (Mohs)	4–4.5
Wavelength of emission maximum (nm)	480
Refractive index	2.2–2.3
Effective average decay time[a] (μs)	13

[a]For γ rays, at room temperature.

Table 2
Samples of $CdWO_4$ crystal scintillators used in this study

ID	Size (mm)	Manufacturer
CWO-1	⌀42 × 39	IIC Novosibirsk[a]
CWO-2	⌀40 × 30	IM Lviv[b]
CWO-3	10 × 10 × 10	ISM Kharkov[c]
CWO-4	⌀25 × 0.9	ISM Kharkov[c]
CWO-5	⌀42 × 25	IM Lviv[b]
CWO-6	10 × 10 × 10	ISM Kharkov[c]
CWO-7	⌀15 × 7	IFNU Lviv[d]

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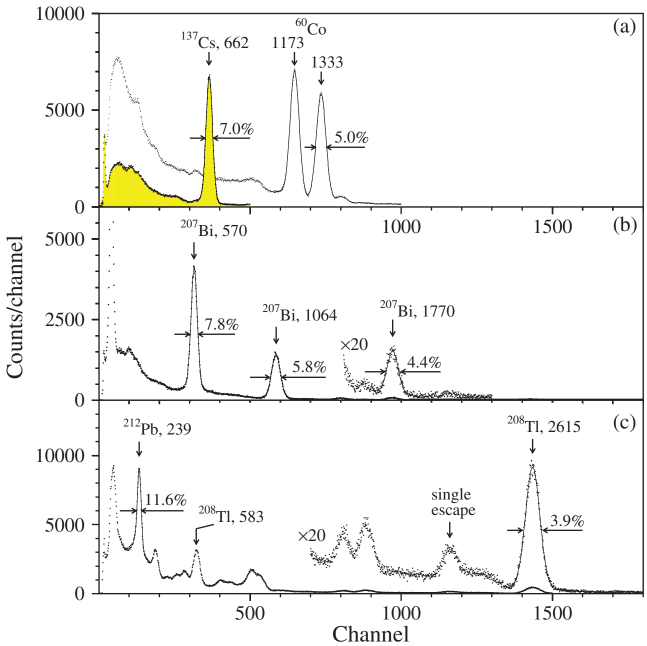

Fig. 1. Energy spectra of ^{60}Co, ^{137}Cs (a), ^{207}Bi (b) and ^{232}Th (c) γ quanta measured by $CdWO_4$ scintillation crystal $\varnothing 42 \times 39$ mm (CWO-1). Energies of γ lines are in keV.

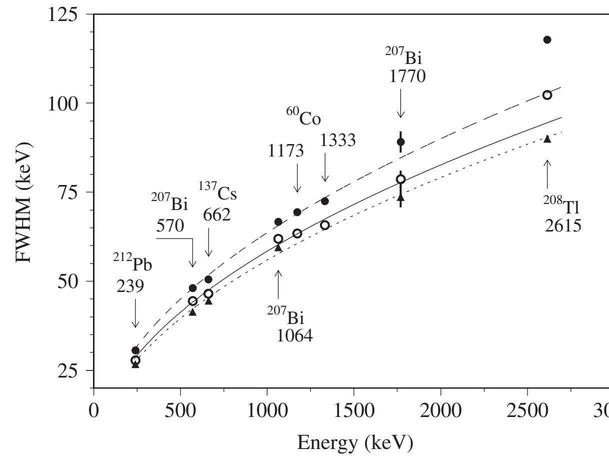

Fig. 2. Dependence of the energy resolution on energy of γ quanta (and their fits by the square root function) measured with CWO-1 (empty circles, solid line), CWO-2 (filled circles, dashed line), and CWO-3 (triangles, dotted line) scintillators.

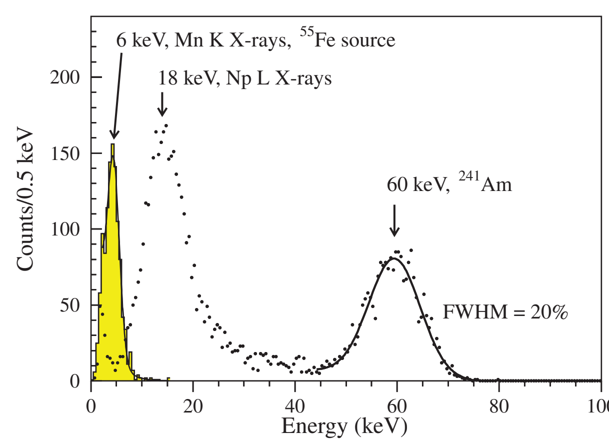

Fig. 3. Energy spectra of ^{55}Fe and ^{241}Am X-rays and γ quanta measured by $CdWO_4$ scintillation crystal $\varnothing 25 \times 0.9$ mm (CWO-4).

with 662, 1064, and 2615 keV γ lines, respectively. In our opinion it is mainly due to the lower transparency of the crystal CWO-2 in comparison with the CWO-1 (see Section 2.3 where the results of measurements of transmittance of the crystals are presented).

Energy resolutions of 6.8%, 5.6% and 3.4% for 662 keV (^{137}Cs), 1064 keV (^{207}Bi), and 2615 keV (^{232}Th) γ lines, respectively, were measured with the small crystal CWO-3.

All these results are summarized in Fig. 2 where the fitting curves are also shown. The square root function with one free parameter was used for the fit: $FWHM = \sqrt{a \times E_\gamma}$, where $FWHM$ is the energy resolution and E_γ is energy of γ quanta in keV. The values $a = 3.40$, 4.12, and 3.07 were obtained for the CWO-1, CWO-2, and CWO-3 crystals, respectively.

Comparable energy resolution with $CdWO_4$ crystal scintillators were obtained in Refs. [12,13,25,30,32].

Light yield of $CdWO_4$ was measured in Ref. [22] with the help of silicon photodiodes as $(15-20) \times 10^3$ photons/MeV (which is $\approx 35-50\%$ of NaI(Tl)). In Ref. [26] a higher photon yield of a cadmium tungstate scintillator ($\approx 28 \times 10^3$ photons/MeV) was estimated on the basis of the measurements reported in Ref. [25]. This result was recently confirmed in Ref. [32], where values in the range $(13-27) \times 10^3$ photons/MeV were reported for $CdWO_4$ crystal scintillators. In Ref. [31] the absolute light yield of $CdWO_4$ scintillators $\approx 20 \times 10^3$ photons/MeV was reported.

The relative photoelectron yield was measured with the CWO-1 crystal and NaI(Tl) $\varnothing 40 \times 40$ mm of standard assembling. Both crystals were coupled to the same PMT XP2412 with the bialkali photocathode and were irradiated by γ quanta of ^{137}Cs source. A transient digitizer based on the Analog Devices 12 bit ADC (AD9022) operated at 20 Mega Sample per second (20 MS/s) [29] was used to accumulate the pulse shapes from the detectors. To build the energy spectra of the $CdWO_4$ and NaI(Tl) scintillators, the area of the pulses was calculated. In such a way we overcome the problem of different decay times of these scintillators. The relative photoelectron yield of the CWO-1 scintillator was measured as 26% of NaI(Tl).

2.1.2. Scintillation response at low energy

Fig. 3 shows the energy spectra of ^{241}Am and ^{55}Fe low energy gamma and X-ray lines measured with thin $CdWO_4$ scintillator $\varnothing 25 \times 0.9$ mm (CWO-4). Even the 6 keV peak of ^{55}Fe is still resolved from the PMT noise. A low energy threshold of a $CdWO_4$ detector is important to search for low energy processes, as for instance, the two neutrino double electron capture in ^{106}Cd. Expected energy release in a crystal in this process is only ≈ 50 keV.

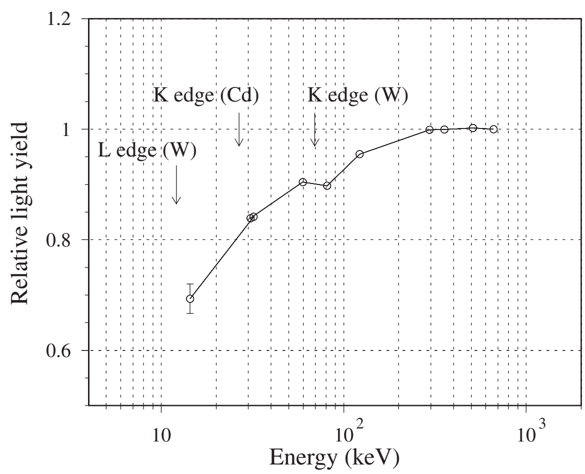

Fig. 4. Non-proportionality in the scintillation response of CWO-4 crystal.

We have studied the non-proportionality in the scintillation response with the CWO-4 scintillator. The crystal was optically connected to EMI9256KB PMT operating at −1000 V. The shaping time of the ORTEC (Model 572) amplifier was set to 10 μs. The γ and X-ray lines from the sources: ^{57}Co (14.4 and 122.1 keV), ^{241}Am (59.5 keV), ^{137}Cs (32.1 and 661.7 keV), ^{133}Ba (30.9, 81.0, 295.3, and 356.0 keV), ^{22}Na (511 keV) were used for the measurements. Positions of the peaks were determined relatively to 661.7 keV γ line of ^{137}Cs. The dependence of the relative photoelectron yield on the energy of X and γ lines is presented in Fig. 4. The behaviour of the scintillator response agrees with the results of other authors [32,35,36]. This effect should be taken into account in experiments to search for low energy processes like, for instance, the neutrino accompanied double electron capture in ^{106}Cd. The energy scale of a detector should be carefully measured in the region of interest.

2.1.3. α/β ratio

Quenching factor for α particles, in other words α/β ratio,[2] is important to interpret and suppress background caused by internal thorium, uranium and α active lanthanides contamination. In Ref. [11] the dependence of the α/β ratio on the energy and direction of α particles relatively to the main crystal axes was observed for $CdWO_4$ crystals. To obtain α particles with energies in the range 0.5–5.3 MeV, a set of thin mylar films (with thickness of 0.65 mg/cm^2) as absorbers were used. The average energies of α particles after the absorbers were measured with the help of a surface-barrier detector. Disadvantage of such an approach is the substantial broadening of the α particles energy after passing the absorbers.

In the present work the 3 MV Tandetron accelerator of the LABEC laboratory of the Sezione di Firenze of INFN [37] was used to obtain beams of alpha particles in the energy range 1–7 MeV. By scattering of the α beam on a thin gold foil energies of α particles of 0.91, 1.86, 2.78, 4.18, and 6.99 MeV were obtained. The CWO-3 crystal was irradiated in the direction perpendicular to the (0 1 0) crystal plane. The obtained dependence of the α/β ratio on the energy of α particles is shown in Fig. 5. The energy spectra measured with 0.91, 2.78, and 6.99 MeV α particles are shown in inset. In the energy interval 2–7 MeV the α/β ratio increases with increasing energy as $\alpha/\beta = 0.093(1) + 0.0173(2)E_\alpha$, where E_α is alpha particle energy in MeV. This result is in agreement with that reported in Ref. [11]. Such a behaviour of the α/β ratio can be explained by the energy dependence of ionization density of α particles [38]. It should be also noted that α/β ratio is not actually a property of a crystal, but more likely a certain characteristics of the detector depending on the shape and surface quality of a crystal, shaping time of electronics, etc.

[2]The α/β ratio is defined as ratio of α peak position in the energy scale measured with γ sources to the energy of α particles.

2.2. Emission spectra

Emission spectra were measured under γ rays (^{60}Co source) and α particles (^{241}Am + ^{239}Pu + ^{241}Cm source) excitation. The $CdWO_4$ crystal, 42 mm in diameter and 25 mm height (CWO-5), was used for the measurements. The fluorescence light was analyzed in wavelength by the SPEX spectrometer. Intensities were integrated over 10 nm intervals. The results of the measurements are presented in Fig. 6a. The emission spectra under γ irradiation are in a good agreement with result reported in Ref. [23]. A small difference in the emission spectra under α particles and γ rays excitation was observed. However, this effect could be due to different absorption of the light emitted by the localized source (α particles) or diffused one (γ quanta).

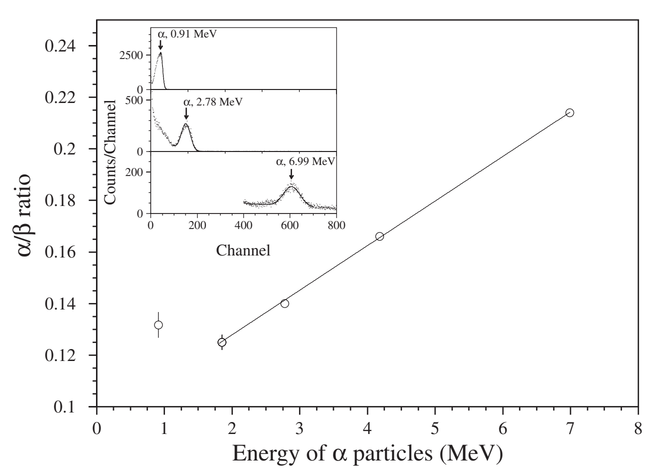

Fig. 5. Dependence of the α/β ratio on energy of α particles measured with CWO-3 crystal with α beam of accelerator. Fit of the data in the energy interval 2–7 MeV by the linear function is shown by solid line. (Inset) The energy spectra measured with 0.91, 2.78, and 6.99 MeV α particles.

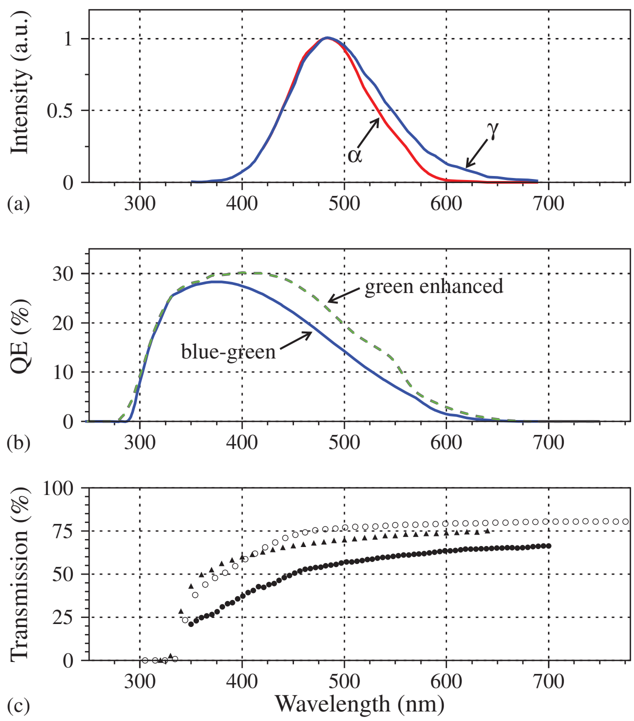

Fig. 6. (a) Emission spectra of CWO-5 crystal excited by γ rays (^{60}Co) and α particles (^{241}Am + ^{239}Pu + ^{241}Cm source). (b) Spectral sensitivity of photomultipliers with blue-green sensitive (PHILIPS, XP2412) and green enhanced bialkali photocathodes (EMI, D724KFL). (c) Optical transmission curve of CWO-1 (empty circles, measured by the producer), CWO-2 (filled circles), and CWO-3 (triangles, measured by the producer) crystals.

It is important to stress that emission spectrum of $CdWO_4$ scintillators is not well fitted to spectral sensitivity of photomultipliers. Value of PMT quantum efficiency to $CdWO_4$ scintillation light can be calculated as the convolution of $CdWO_4$ emission spectrum and spectral sensitivity of the PMT photocathode. We have obtained $QE = 0.13$ using the measured emission spectrum of $CdWO_4$ (see Fig. 6a) and specification of the PMT (XP2412) with bialkali (blue-green sensitive) photocathode presented in Fig. 6b. For the high quality PMT with green-enhanced (RbCs) photocathode (EMI D724KFL, serial #13, see Fig. 6b) produced by THORN EMI for the Solotvina experiment [9], we have obtained the value $QE = 0.17$.

2.3. Light transmission and scattering

Transmittance of the CWO-2 crystal was measured in the spectral range 350–700 nm with the help of the spectrophotomether KSVU-23 equipped with reflection attachment. The transmission curve is shown in Fig. 6c (filled circles). Taking into account the reflection losses, the value of ≈ 10–15 cm for attenuation length of the $CdWO_4$ crystal was obtained at the maximum of emission spectra (485 nm). Transmittance of the CWO-1 and CWO-3 crystals measured by the producers is also presented in Fig. 6c. The crystals CWO-1 and CWO-3 show much better optical properties, namely the value of attenuation length is ≈ 50–70 cm at the wavelength of the maximum of the emission spectrum.

Generally speaking, light attenuation in crystals is caused by absorption and scattering. The angular dependence of light intensity after passing the CWO-2 crystal was measured to estimate the light scattering in the crystal. Fig. 7 shows the layout of the measurement. A laser beam (expansion angle less than 1 mrad) of 632.8 nm wavelength and 0.5 mm diameter was used. The beam was directed normally to the face of the crystal. Intensity of the beam was measured by a Si-photodetector with diameter of 11 mm. The distance l between the photodetector and the crystal was varied in the range 30–1350 mm.

The measured dependence on the solid angle Ω (Fig. 7) is well described by logarithmic function, and shows a considerable forward light scattering in the $CdWO_4$ crystal. No dependence was observed in the measurements without crystal, as well as with the 30 mm-thick optical glass (K-8) installed instead of the crystal. The observed behaviour of light scattering can be explained by substantial amount of optical inhomogeneities whose sizes are comparable or exceed wavelength of the light [39]. Non-stoichiometric composition, presence of regions with distorted (or disturbed) structure, especially with partially amorphous structure, pores, voids, flaws, inclusions, can be causes of these inhomogeneities in $CdWO_4$ crystals.

Processes of light scattering should be taken into account in simulation of light collection in $CdWO_4$ scintillation detectors.

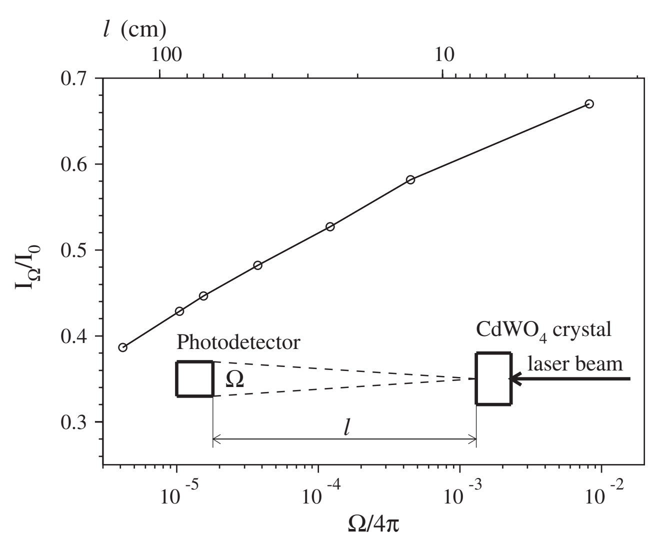

Fig. 7. Dependence of the I_Ω/I_0 ratio on the solid angle Ω under which the beam of the laser (passing through $CdWO_4$ crystal) reaches the photodetector. Here I_Ω is the measured light intensity within the solid angle Ω, and I_0 is the incident light intensity. The schematic view of the set up is also shown.

2.4. Scintillation decay

2.4.1. Pulse shape for γ rays and α particles

Pulse shapes of $CdWO_4$ scintillators were studied as described in Refs. [29,11] with the help of a transient digitizer based on the 12 bit ADC (AD9022) operated at 20 MS/s. However, the integration time of the preamplifier in the present measurements was decreased ($\approx 0.02\,\mu s$ in comparison with $\approx 0.2\,\mu s$ in Refs. [29,11]) to investigate possible fast components of scintillation decay. More recently, pulse shape for γ rays and α particles was measured also with the help of the 12 bit 125 MS/s transient digitizer described in Refs. [40,41]. Furthermore, single-electron counting method was applied to study the dependence of $CdWO_4$ scintillation signal for α particles and γ quanta on emission wavelength (see Section 2.4.4).

To study pulse shape of scintillation decay for α particles, the $CdWO_4$ crystal 10 × 10 × 10 mm (CWO-6) was irradiated by α particles from collimated ^{241}Am source in the direction perpendicular to the (0 1 0) crystal plane. The dimensions of the collimator were ∅0.75 × 2 mm. The energy of α particles after passing of 2 mm air layer was calculated by GEANT3.4 program as 5.25 MeV [11]. ^{60}Co was used as a source of γ quanta. Measurements were carried out at room temperature (23 ± 2) °C.

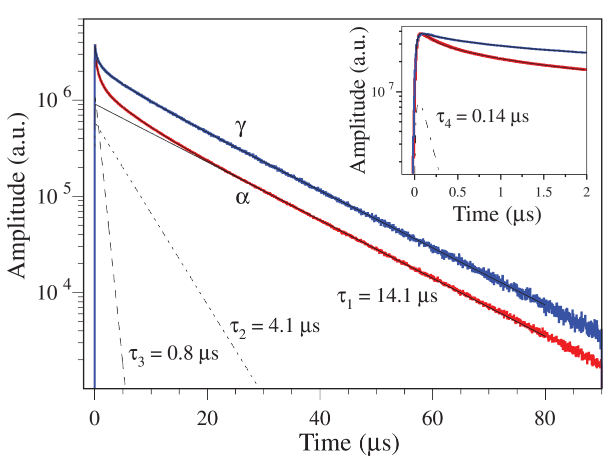

Fig. 8. Decay of scintillation in CWO-6 crystal for γ rays and α particles measured by 20 MS/s transient digitizer. (Inset) The first part of the pulses measured with 125 MS/s digitizer. Four components of scintillation signal from α particles with time decay of 0.14, 0.8, 4.1 and 14.1 μs are shown. Fitting functions for α and γ pulses are shown by solid lines.

The shapes of the light pulses produced by α particles and γ rays in the $CdWO_4$ scintillator measured by the 20 MS/s digitizer are shown in Fig. 8. To obtain the pulse shapes, large numbers of individual α and γ events (with amplitudes corresponding to α peak of ^{241}Am) were summed. The first part of $CdWO_4$ α and γ pulses measured with the help of the 125 MS/s digitizer is presented in the inset of Fig. 8. A fit to the pulses was done by the function:

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

where A_i are the relative intensities, τ_i—the decay constants for different light-emission components, and τ_0 is integration constant of electronics ($\tau_0 \approx 0.02\,\mu s$). Four decay components were observed with $\tau_i \approx 0.1-0.2\,\mu s$, $\approx 1\,\mu s$, $\approx 4\,\mu s$ and $\approx 14-15\,\mu s$ with different intensities for γ rays and α particles (see Table 3). Similar results have been obtained with the crystal CWO-7 studied both with the 20 and 125 MS/s digitizers.

2.4.2. Pulse-shape discrimination between γ rays and α particles

The difference of the pulse shapes allows to discriminate γ(β) events from those induced by α particles. We applied for this purpose the optimal filter method proposed in Ref. [42] and already applied to $CdWO_4$ scintilators in Ref. [29]. For each $CdWO_4$ signal a numerical parameter (shape indicator, SI) was calculated in the following way:

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

where the sum is over time channels k, starting from the origin of pulse and up to certain time (75 μs for 20 MS/s digitizer and 64 μs for 125 MS/s), $f(t_k)$ is the digitized amplitude (at the time t_k) of the signal. The weight function $P(t)$ was 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.

Clear discrimination between α particles and γ rays was achieved using this approach, as one can see in Fig. 9 where the SI distributions measured by the 125 MS/s transient digitizer with the CWO-7 scintillation crystal for α particles ($E_\alpha \approx 5.3$ MeV) and γ quanta (≈ 1.2 MeV) are shown. As a

Table 3
Decay time of $CdWO_4$ scintillators for γ quanta and α particles measured by transient digitizers at room temperature

Type of irradiation	Decay constants (μs) and relative intensities			
	τ_1 (A_1)	τ_2 (A_2)	τ_3 (A_3)	τ_4 (A_4)
α particles	14.1 ± 0.3 (79.2 ± 2.0)%	4.1 ± 0.6 (14.5 ± 1.2)%	0.8 ± 0.2 (5.0 ± 0.6)%	0.14 ± 0.07 (1.3 ± 0.4)%
γ rays	14.5 ± 0.3 (88.7 ± 2.0)%	4.6 ± 0.8 (8.7 ± 1.5)%	0.8 ± 0.2 (2.1 ± 0.4)%	0.15 ± 0.05 (0.5 ± 0.2)%

The decay constants and their relative intensities are denoted as τ_i and A_i, respectively.

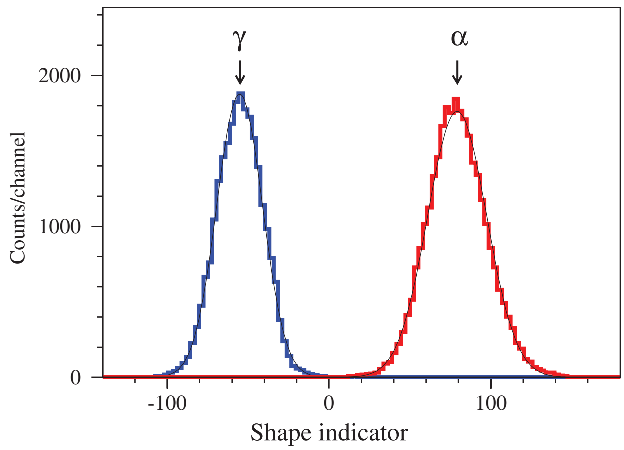

Fig. 9. The shape indicator (see text) distributions measured by CWO-7 detector with α particles ($E_\alpha = 5.3$ MeV) and γ quanta (≈ 1.2 MeV) using the 125 MS/s 12 bit transient digitizer. The distributions were fitted by Gaussian function (solid lines).

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

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

where SI_α and SI_γ are mean SI values for α particles and γ quanta distributions (which are well described by Gaussian functions, see Fig. 9), σ_α and σ_γ are the corresponding standard deviations. For the distributions presented in Fig. 9, the factor of merit is FOM = 5.8. This value is slightly better than that of FOM = 5.6 obtained by using the 20 MS/s transient digitizer.

2.4.3. Pulse shape and fluorescence light wavelength under laser excitation

Measurements with pulses of ultraviolet light have been performed in order to investigate whether the fluorescence emission contains at least part of the components of different lifetime observed in α particles and γ induced scintillation, and to search for the possible dependence of pulse shape on the wavelength of the emitted light. This part of the work has been performed at the European Laboratory for non-linear Spectroscopy (LENS, Florence).

The fluorescence of the CWO-7 crystal has been excited by fast ultraviolet pulses from a laser source (λ = 266 nm), and the time dependence of the emitted light has been investigated in different intervals of wavelength, of 10 nm width, centered at 380, 440, 470, 500, 560, 600, and 650 nm [43]. In the experimental set-up the 1064 nm light from a YAG:Nd laser was used to excite a pair of non-linear crystals tuned to generate the fourth harmonics. The resulting 266 nm radiation was focused on the face of the $CdWO_4$ crystal. The fluorescence light, analyzed in wavelength by the SPEX Spectrometer (22 cm focal length), was collected by an EMI9813 PMT, which was located close to the exit slits of the Spectrometer. The pulses from the anode of the PMT were integrated with a time constant of ≈ 0.2 μs, and sent to the input of a digital

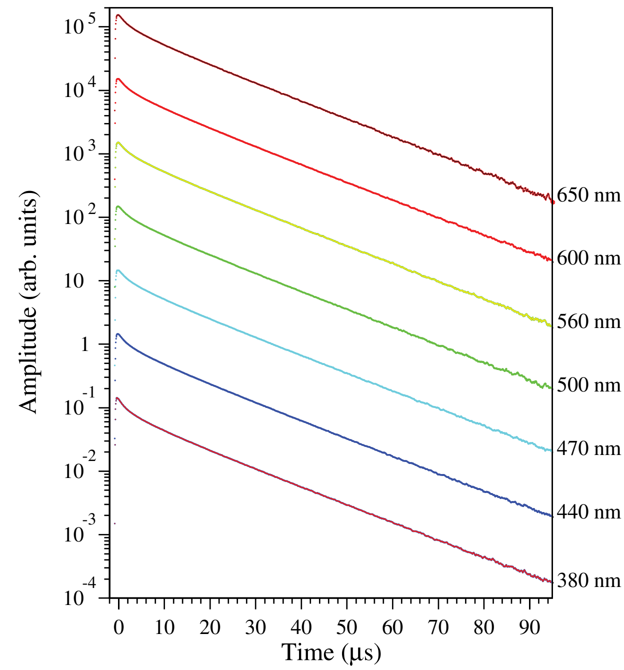

Fig. 10. Pulse shape of the fluorescence light for different intervals of wavelengths (20 nm wide). Shapes are normalized to equal area and shifted by a decade to improve visibility.

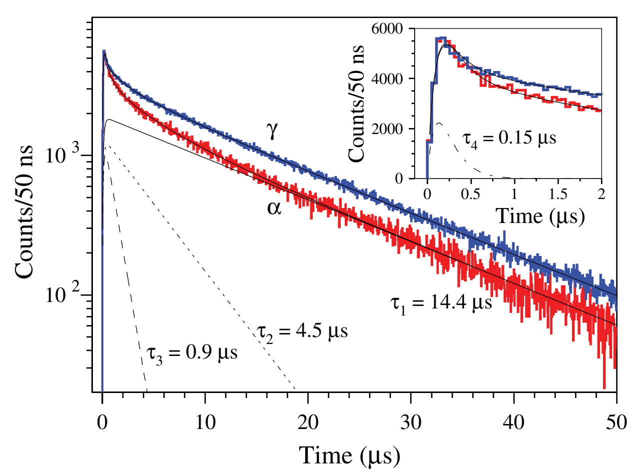

Fig. 11. Decay of scintillation in CWO-7 crystal for γ rays and α particles measured by single electron counting method. The scintillation light was passing through interference filter with central wavelength of 480 nm (see text). The first part of the pulses is shown in inset. Four components of α scintillation signal with time decay of 0.15, 0.9, 4.5 and 14.4 μs are shown. Fitting functions for α and γ pulses are shown by solid lines.

oscilloscope (HP TDS460). The digital output of the oscilloscope was transmitted to a computer and stored in memory for further analysis.

The pulse shapes (corresponding to the average of a large number of individual pulses) of the $CdWO_4$ fluorescence light with the different wavelengths are shown in Fig. 10. Three components of the scintillation decay with decay times and intensities $\tau_1 \approx 15$ μs (85%), $\tau_2 \approx 5$ μs (11%), and $\tau_3 \approx 1$ μs (4%) were observed. We were not able to measure the fast $\approx 0.1-0.2\,\mu s$ decay component found in our measurements with the digitizers and by using single electron counting method, because of the rather big integration constant used in the measurements with laser excitation.

No dependence of pulse shape on the wavelength of emitted light under the laser excitation was observed.

2.4.4. Study of scintillation decay time for α particles and γ quanta at different wavelength of emission spectra

The pulse shape for α particles and γ quanta at different wavelength were measured by the single photon counting method. The CWO-7 crystal scintillator was optically connected to EMI9256KB PMT. The signal from the PMT gives the start signal to the time-digital converter (Time Analyzer, Canberra, Model 2143). Scintillation light from the $CdWO_4$ crystal entered through the diaphragm 10 mm in diameter and passed through interference filters (Edmund Scientific Co.) with central wavelength 420, 460, 480, 590 nm to a PMT cooled down to −20 °C (Product for Research, inc, USA). The PMT operating at single electron counting mode generated stop signals for the converter. The time scale of the time-digital converter was calibrated with the help of an ORTEC Model 462 Time Calibrator.

The pulse shapes of $CdWO_4$ scintillator for α particles ($^{241}Am + ^{239}Pu + ^{241}Cm$ source) and γ quanta (^{137}Cs)

Table 4
Decay time of CWO-7 scintillator for γ quanta and α particles measured by single electron counting method at room temperature with 480 nm filter (see text)

Type of irradiation	Decay constants (μs) and relative intensities			
	τ_1 (A_1)	τ_2 (A_2)	τ_3 (A_3)	τ_4 (A_4)
α particles	14.4 ± 0.5 (78.3 ± 5.0)%	4.5 ± 0.7 (16.0 ± 4.0)%	0.9 ± 0.2 (4.4 ± 0.6)%	0.15 ± 0.03 (1.3 ± 0.4)%
γ rays	14.7 ± 0.2 (89.0 ± 2.0)%	4.2 ± 0.4 (8.4 ± 1.0)%	0.9 ± 0.2 (1.9 ± 0.4)%	0.11 ± 0.04 (0.7 ± 0.2)%

The decay constants and their relative intensities are denoted as τ_i and A_i, respectively.

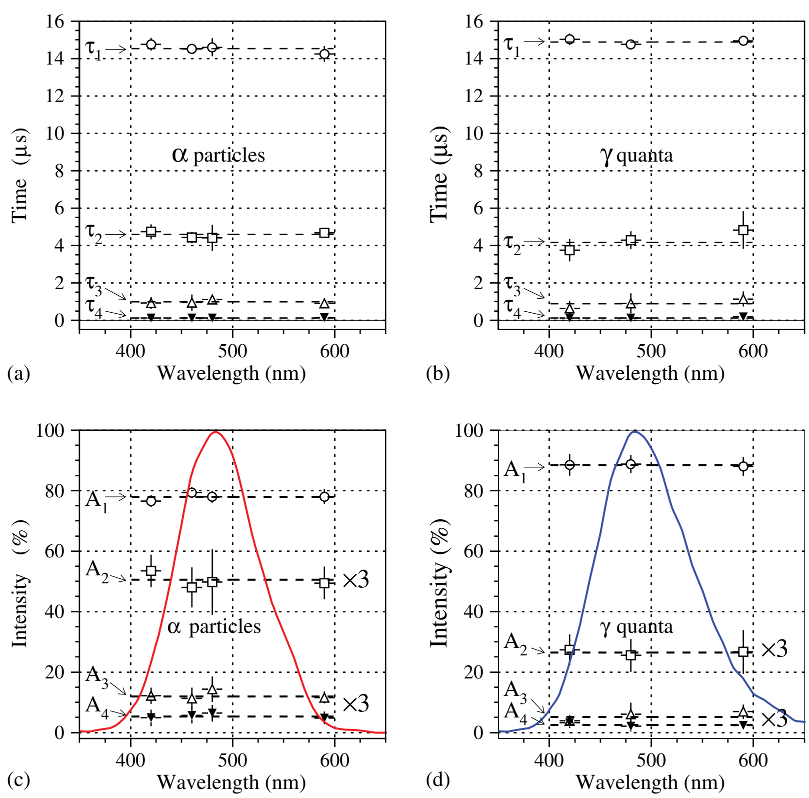

Fig. 12. (a,b) The time constants (τ_i) and (c,d) intensities (A_i) of $CdWO_4$ (CWO-7) scintillation signals for α particles and γ quanta measured by single electron counting method for different emission wavelengths. Data for A_2, A_3, and A_4 are multiplied by a factor of 3 to improve visibility. The measured emission spectra of $CdWO_4$ crystal (CWO-5) excited by α particles (c) and γ rays (d) are also drawn.

measured by the single electron counting method with the 480 nm filter are depicted in Fig. 11. Fit of the obtained forms by sum of four exponential components gives values of the decay constants and their intensities (Table 4) similar to that obtained with the help of the transient digitizers. The results of the measurements with the different filters are presented in Fig. 12. No dependence of decay times on wavelength of the emission spectra both for α particles and γ quanta was observed.

According to Ref. [19], the spectral composition of the light emitted by $CdWO_4$ should contain two different parts, one in the blue-green region, the other in the yellow region. In our measurements the latter can hardly be recognized over the tail of the blue-green component. The time distribution of the emitted light does not change significantly in the wavelength region from 380 to 650 nm under laser excitation nor under α and γ irradiation in the region of 420–590 nm.

2.4.5. Dependence of pulse shape on temperature

Temperature dependence of the pulse shape for γ rays and α particles was checked in the range 0–25 °C. The CWO-4 crystal was optically connected to EMI9256KB PMT operating at −1000 V. The scintillation crystal and the PMT were kept at the same temperature. The pulse shape was recorded by the 12 bit 20 MS/s transient digitizer. The crystal was irradiated by γ rays from ^{60}Co source and α particles from ^{241}Am source. Forms of signals for γ rays and α particles have been obtained as a result of summation of several 1000 individual pulses. The values of the time constants and their intensities were obtained by fitting of the forms. The sum of four exponential functions has been taken as model for the description of scintillation signals.

Temperature dependence of the decay time constants and their intensities are presented in Fig. 13. The decay component τ_1 depends on the temperature as −0.055(3) μs/°C for α particles and as −0.048(3) μs/°C for γ quanta. It should be noted that the intensities of this component both for α and γ signals remain constant: (77.8 ± 0.2)% for α particles and (88.8 ± 0.2)% for γ quanta.

The temperature dependence of the averaged decay time is shown in Fig. 14a. The averaged decay time decrease with temperature as ≈ −0.050(4) μs/°C for α particles and ≈ −0.048(7) μs/°C for γ quanta, which is in an agreement with results reported by Melcher et al. [23]. This dependence is mainly due to the temperature dependence of the τ_1 decay component.

The factor of merit of pulse-shape discrimination between α particles and γ quanta was calculated for the data accumulated in the temperature interval of 0–24 °C.

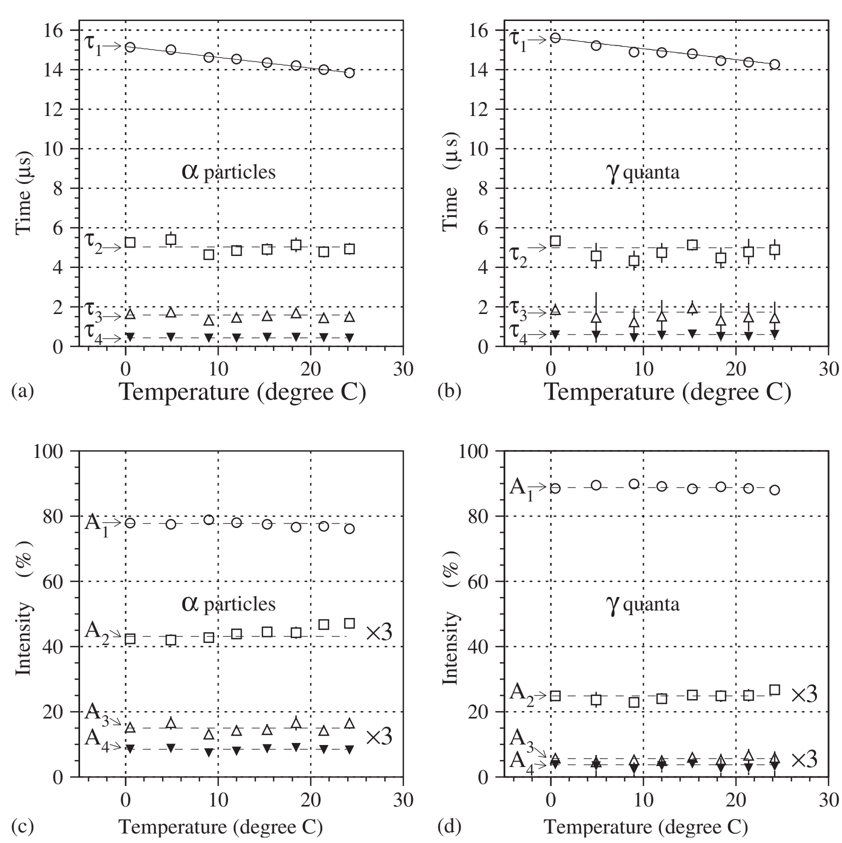

Fig. 13. (a, b) Temperature dependence of the time constants (τ_i) and (c, d) intensities (A_i) of $CdWO_4$ scintillation signals for α particles and γ quanta measured with the crystal CWO-4. Data for A_2, A_3, and A_4 are multiplied by a factor of 3 to improve visibility. The results of fit by the linear function (time constants τ_1) and by constants (τ_2, τ_3, τ_4, and all intensities) are shown.

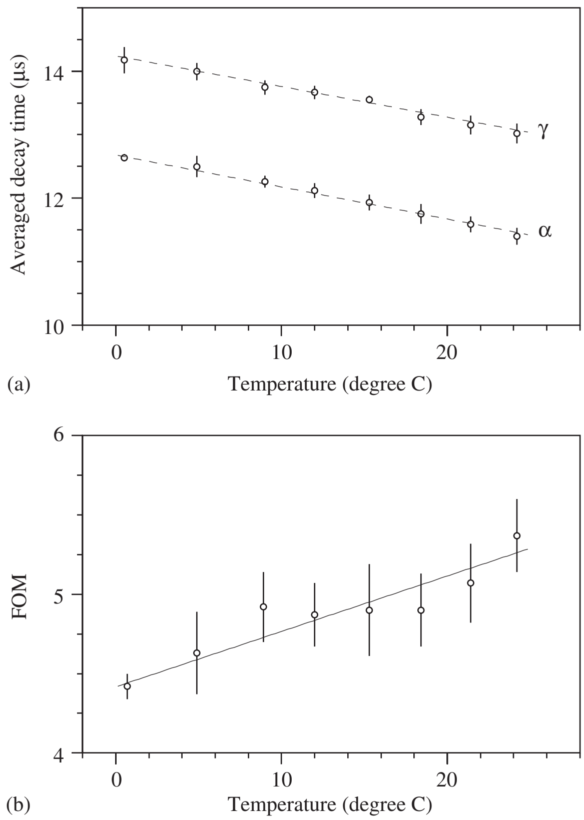

Fig. 14. (a) Temperature dependence of the averaged scintillation decay time for γ quanta (≈ 1.2 MeV) and α particles (≈ 5.3 MeV) in CWO-4 crystal. (b) The pulse-shape discrimination efficiency (denoted as FOM—"factor of merit", see text) is slightly improved with increasing of the temperature.

The weight function (see Section 2.4.2) was constructed by using pulse shapes for α particles and γ quanta measured at room temperature. As one can see in Fig. 14b, the factor of merit is slightly improved with increase of temperature. It could be explained by the increase of the difference between scintillation decay times under α and γ excitation with increasing of temperature.

3. Conclusions

Scintillation properties of $CdWO_4$ crystals were studied. The energy resolution 7.0% and 3.9% for the 662 and 2615 keV γ lines was obtained with large (∅42 × 39 mm) $CdWO_4$ crystal scintillator. Small crystal (10 × 10× 10 mm) showed an even better energy resolution: 6.8% and 3.4% for the 662 and 2615 keV γ lines, respectively.

Spectra of the low energy γ and X-ray lines (6 keV of ^{55}Fe, 18 keV of Neptunium L line and 60 keV γ quanta from ^{241}Am source) were measured, which demonstrates possibility to apply $CdWO_4$ crystal scintillators to search for double electron capture in ^{106}Cd. Non-proportionality in the scintillation response observed in the present work is in agreement with that reported by other authors.

The energy dependence of the α/β ratio was measured with α beam produced by accelerator. Behaviour of the dependence is in an agreement with that reported in Ref. [11]. The α/β ratio increases linearly in the energy interval 2–7 MeV.

A difference in long-wavelength part of the emission spectra for γ rays and α particles was observed, however we cannot exclude that this effect is due to different absorption of scintillation light emitted under γ and α irradiation.

Transmissivity of $CdWO_4$ crystals was measured and considerable scattering of light was observed. These data indicate a presence of a substantial amount in our $CdWO_4$ crystal of optical heterogeneity whose sizes are comparable or exceed the scintillation light wavelength.

Four components of scintillation decay ($\tau_i \approx 0.1-0.2$, ≈ 1, ≈ 4 and $\approx 14-15\,\mu s$) and their intensities under α particles and γ quanta irradiation were measured with different $CdWO_4$ crystal scintillators by using different methods: transient digitizers with 20 and 125 MHz sampling frequency as well as single electron counting method. The difference in the scintillation pulse shapes for α particles and γ quanta is mainly due to difference in the intensities of the different decay components.

Clear discrimination between α particles and γ rays was achieved using the optimal filter method.

No dependence of the pulse shape of the $CdWO_4$ fluorescence light on wavelengths was observed in the range 380–650 nm under laser excitation as well as under α particles and γ quanta irradiation in the range 420–590 nm.

Temperature dependence of the decay constants and intensities of $CdWO_4$ pulse shape for γ rays and α particles was investigated in the temperature range of 0–24 °C. Clear temperature dependence of the $\approx 14-15\,\mu s$ component at the level of $\approx -0.05\,\mu s/^{\circ}C$ was observed for α particles and γ quanta, while the intensities of this component both for α and γ signals remain constant. The pulse-shape discrimination improved slightly with increasing of temperature.

Acknowledgments

It is very pleasant to express gratitude to the personnel of the LABEC laboratory of the Sezione di Firenze of INFN, Prof. P.A. Mando, Dr. M. Chiari, Dr. L. Giuntini for the opportunity to carry out the measurements with a beam of alpha particles. The authors would like to thank Prof. M. Pashkovskii and Dr. M. Batenchuk from the Ivan Franko National University (Lviv, Ukraine) for providing one of the $CdWO_4$ crystals used in the present study. We are grateful to Prof. A. Vinattieri from the Physics Department of Florence University for lending of the interference filters and the device for the single electron counting measurements.

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