Further study of CdWO₄ crystal scintillators as detectors for high sensitivity 2β experiments: scintillation properties and pulse-shape discrimination

L. Bardelli⁺, M. Bini⁺, P.G. Bizzeti⁺, L. Carraresi⁺, F.A. Danevich⁰, T.F. Fazzini⁺, B.V. Grinyov⁺, N.V. Ivanikova⁴, V.V. Kobychev⁰, B.N. Kropivynsky⁰, P.R. Maurenzig⁺, L.L. Nagornaya⁰, S.S. Nagorny⁴, A.S. Nikolaiko⁰, A.A. Pavlyuk⁴, D.V. Poda⁰, I.M. Solsky⁺, M.V. Sopinsky⁺, Yu.G. Stenin⁴, F. Taccetti⁺, V.I. Tretyak⁰, Ya.V. Vasiliev⁴, S.S. Yurchenko⁰

⁺ Dipartimento di Fisica, Università di Firenze and INFN, 50019 Firenze, Italy
⁰ Institute for Nuclear Research, MSP 03680 Kiev, Ukraine
⁴ Institute for Scintillation Materials, 61001 Kharkov, Ukraine
⁺ Institute for Nuclear Research, MSP 03680 Kiev, Ukraine
⁶ Institute for Materials, 79031 Lviv, Ukraine
⁷ Lashkaryov Institute of Semiconductor Physics, 03028 Kiev, Ukraine

Abstract

Energy resolution, light yield, non-proportionality in the scintillation response, α/β ratio, pulse shape for γ rays and α particles were studied with CdWO₄ crystal scintillators. Some indication for a difference in the emission spectra for γ rays and α particles was observed. No dependence of CdWO₄ 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.

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]). 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 (⁴¹⁶CdWO₄) crystal scintillators [8, 9]. The half-life limit on 0ν2β decay of ¹¹⁶Cd was set as \( T_{1/2} \geq 1.7 \times 10^{23} \) yr at 90% C.L., which corresponds to an upper bound on the effective Majorana neutrino mass \( \langle m_\nu \rangle \leq 1.7 \) 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 ⁷⁶Ge, ⁸²Se, ⁱ⁰⁰Mo, ¹³⁰Te, and ¹³⁶Xe).

Two by-product results obtained in the course of the Solotvina experiment with CdWO₄ 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 ¹¹³Cd were measured [10];

An evidence for 0ν2β decay of ⁷⁶Ge has been claimed in [2]. However, it was criticized in [3, 4, 5]. Later the Heidelberg group has presented new data with improved statistics and after a reanalysis. A half-life \( T_{1/2} \approx 1.2 \times 10^{25} \) y has been reported [9], which corresponds to the effective Majorana neutrino mass \( \langle m_\nu \rangle \approx 0.4 \) eV.
(ii) indication for the $\alpha$ 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 $2\beta$ 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 $\approx 100$ 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 $2\beta$ decay experiment with $^{116}$CdWO$_4$ by using lead tungstate crystal scintillators as a high efficiency $4\pi$ active shield [13].

In addition, as it was demonstrated in [14], CdWO$_4$ crystals show good potential to develop thermal bolometers with energy resolution $\approx 5$ 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 $2\beta$ experiment: energy resolution, light yield, non-proportionality in the scintillation response, $\alpha/\beta$ ratio, emission spectra and transparency, pulse shape for $\gamma$ rays and $\alpha$ particles and their temperature dependence.

2 MEASUREMENTS AND RESULTS

The luminescence of CdWO$_4$ crystals was discovered about sixty years ago [16]. The different properties of CdWO$_4$ scintillators were investigated (see Refs. [17, 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32] and references therein). In 1960 G.B. Beard and W.H. 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].
Table 1: Properties of CdWO₄ crystal scintillators

| Property                                      | Value     |
|----------------------------------------------|-----------|
| Density (g/cm³)                              | 7.9       |
| Melting point (°C)                           | 1271      |
| Structural type                              | Wolframite|
| Cleavage plane                               | Marked (010) |
| Hardness (Mohs)                              | 4 – 4.5   |
| Wavelength of emission maximum (nm)          | 480       |
| Refractive index                             | 2.2 – 2.3 |
| Effective average decay time* (µs)           | 13        |

*For γ rays, at room temperature.

Table 2: Samples of CdWO₄ crystal scintillators used in this study

| ID  | Size (mm) | Manufacturer                       |
|-----|-----------|------------------------------------|
| CWO–1| ⊙42 × 39 | IIC Novosibirsk<sup>a</sup>         |
| CWO–2| ⊙40 × 30 | IM Lviv<sup>b</sup>                |
| CWO–3| 10 × 10 × 10 | ISM Kharkov<sup>c</sup>        |
| CWO–4| ⊙25 × 0.9 | ISM Kharkov<sup>c</sup>           |
| CWO–5| ⊙42 × 25 | IM Lviv<sup>b</sup>               |
| CWO–6| 10 × 10 × 10 | ISM Kharkov<sup>c</sup>        |
| CWO–7| ⊙15 × 7 | IFNU Lviv<sup>d</sup>             |

<sup>a</sup> Nikolaev Institute of Inorganic Chemistry, Novosibirsk, Russia
<sup>b</sup> Institute for Materials, Lviv, Ukraine
<sup>c</sup> Institute for Scintillation Materials, Kharkov, Ukraine
<sup>d</sup> Ivan Franko National University, Lviv, Ukraine
2.1 Scintillation properties

2.1.1 Energy resolution

In the present work, the energy resolution was measured with three CdWO₄ crystals: \( \varnothing 42 \times 39 \text{ mm} \) (CWO–1), \( \varnothing 40 \times 30 \text{ mm} \) (CWO–2), and \( 10 \times 10 \times 10 \text{ mm} \) (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 \( \mu \text{s} \) shaping time to collect most of the charge from the anode of the PMT. The detector was irradiated by \( \gamma \) quanta of \(^{60}\text{Co}, ^{137}\text{Cs}, ^{207}\text{Bi}, \) and \(^{232}\text{Th} \) sources. The energy resolutions (full width at half maximum, FWHM) of 7.0\% (\(^{137}\text{Cs}, 662 \text{ keV}\)), 5.8\% (\(^{207}\text{Bi}, 1064 \text{ keV}\)), 5.0\% (\(^{60}\text{Co}, 1333 \text{ keV}\)), and 3.9\% (\(^{232}\text{Th}, 2615 \text{ keV}\)) were obtained (see Fig. 1).

![Figure 1: Energy spectra of \(^{60}\text{Co}, ^{137}\text{Cs} \), \(^{207}\text{Bi} \) and \(^{232}\text{Th} \) \( \gamma \) quanta measured by CdWO₄ scintillation crystal \( \varnothing 42 \times 39 \text{ mm} \) (CWO–1). Energies of \( \gamma \) lines are in keV.](image)

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 with 662, 1064, and 2615 keV \( \gamma \) 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 subsection 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 (\(^{137}\text{Cs}\)), 1064 keV (\(^{207}\text{Bi}\)) and 2615 keV (\(^{232}\text{Th}\)) \( \gamma \) 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 $E_\gamma$ is energy of $\gamma$ 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.

Figure 2: Dependence of the energy resolution on energy of $\gamma$ 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.

Comparable energy resolution with CdWO$_4$ crystal scintillators were obtained in [25, 12, 30, 32, 13].

2.1.2 Light yield

Light yield of CdWO$_4$ was measured in [22] with the help of silicon photodiodes as $(15 - 20) \times 10^3$ photons/MeV (which is $\approx 35 - 50\%$ of NaI(Tl)). In [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 [25]. This result was recently confirmed in [32], where values in the range $(13 - 27) \times 10^3$ photons/MeV were reported for CdWO$_4$ crystal scintillators. In [31] the absolute light yield of CdWO$_4$ scintillators $\approx 20 \times 10^3$ photons per MeV was reported.

We try to estimate an absolute light yield of CdWO$_4$ crystal scintillators by using data of energy resolution measurements. For an ideal scintillation detector the energy resolution for $\gamma$ rays is given by [35]:

$$\frac{FWHM}{E} = \frac{2.355}{\sqrt{\bar{N}_{phe}}} \times 100\%,$$

(1)

where $\bar{N}_{phe}$ is the mean number of photoelectrons produced in photocathode of PMT. The number of photoelectrons can be written as product:
\[ N_{phe} = N_{ph} \times E_\gamma \times LC \times QE, \]  

where \( N_{ph} \) is mean number of photons created in a scintillator per 1 MeV of energy deposit, \( E_\gamma \) – the energy of \( \gamma \) quanta in MeV, \( LC \) - the fraction of scintillation photons arrived to the photocathode of PMT, \( QE \) – the quantum efficiency of the PMT photocathode to photons emitted by the scintillator.

Value of \( QE \) can be calculated as the convolution of \( \text{CdWO}_4 \) emission spectrum and spectral sensitivity of the PMT photocathode. We have obtained \( QE = 0.13 \) using the measured emission spectrum of \( \text{CdWO}_4 \) (see subsection 2.2 and Fig. 6, a) and specification of the PMT (XP2412) with bialkali (blue-green sensitive) photocathode. For the high quality PMT with green-enhanced (RbCs) photocathode (EMI D724KFL, serial #13) produced by THORN EMI for the Solotvina experiment, we have obtained the value \( QE = 0.17 \).

To estimate the value of \( LC \), light propagation in the \( \text{CdWO}_2 \) detector was Monte Carlo simulated with the help of the GEANT4 package. The emission spectrum and optical transmission curve of the \( \text{CdWO}_4 \) crystal (see subsection 2.3), and the spectral sensitivity of the PMT photocathode were taken into account. An overall light collection of 27% was calculated. Such a modest value is mainly due to absorption of light and large refractive index of \( \text{CdWO}_4 \) (2.2 – 2.3).

Using formulas 1 and 2, an absolute light yield in the range \((30 - 41) \times 10^3\) photons/MeV was calculated for the \( \text{CdWO}_4 \) scintillation crystal. At least the lower border of this estimation is in agreement with the results reported in [26, 32].

The absolute photon yield was also estimated with the help of the \( \text{CWO–3} \) crystal scintillator. The energy resolution was measured in different conditions of light collection. However, we select a geometry (the polished \( \text{CWO–3} \) crystal without reflector, covered by black cope, optically coupled to the PMT) which can be simulated with a comparatively high degree of accuracy. In this case we do not need to simulate diffused surfaces, light propagation from crystal with further reflection and return into scintillator, etc. The energy resolution of \( 8.5 \pm 0.3\% \) was measured for 662 keV \( \gamma \) quanta of \( ^{137}\text{Cs} \), while the value of the light collection for this detector was calculated as 23%. The photon yield was estimated to be of \( 41 \times 10^3 \) photons per 1 MeV of energy deposit, which is more than that reported in [26, 31, 32]. At the same time we realize that the main systematic error in the estimations of absolute light yield can be due to not quite correct calculations of the light collection. In our opinion further investigations are necessary to determine the absolute light yield of \( \text{CdWO}_4 \) scintillators.

The relative photoelectron yield was measured with the \( \text{CWO–1} \) crystal and NaI(Tl) \( \odot 40 \times 40 \) mm of standard assembling. Both crystals were coupled to the same PMT XP2412 with the bialkali photocathode and were irradiated by \( \gamma \) quanta of \( ^{137}\text{Cs} \) source. A transient digitizer based on the Analog Devices 12 bit ADC (AD9022) operated at 20 Mega Sample per second (20 MS/s) was used to accumulate the pulse shapes from the detectors. To build the energy spectra of the \( \text{CdWO}_4 \) 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 \( \text{CWO–1} \) scintillator was measured as 26% of NaI(Tl).
2.1.3 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 $\odot 25 \times 0.9$ 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 $\approx 50$ keV.

![Energy spectra](image)

Figure 3: Energy spectra of $^{55}$Fe and $^{241}$Am X-rays and $\gamma$ quanta measured by CdWO$_4$ scintillation crystal $\odot 25 \times 0.9$ mm (CWO–4).

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 volts. The shaping time of the ORTEC (Model 572) amplifier was set to 10 $\mu$s. The $\gamma$ and X ray lines from the sources: $^{57}$Co (14.4 and 122.1 keV), $^{241}$Am (17.6 and 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 $\gamma$ line of $^{137}$Cs. The dependence of the relative photoelectron yield on the energy of X and $\gamma$ lines is presented in Fig. 4. The behaviour of the scintillator response agrees with the results of other authors [37, 38]. 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.
2.1.4 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 [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 [39] 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 (010) 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 \(E_\alpha\) is alpha particle energy in MeV. This result is in agreement with that reported in [11]. Such a behaviour of the α/β ratio can be explained by the energy dependence of ionization density of α particles [11]. 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.
Figure 5: Dependence of the $\alpha/\beta$ ratio on energy of $\alpha$ particles measured with CWO–3 crystal with $\alpha$ 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 $\alpha$ particles.

2.2 Emission spectra

Emission spectra were measured under $\gamma$ rays ($^{60}$Co source) and $\alpha$ 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. 6, a. The emission spectra under $\gamma$ irradiation are in a good agreement with result reported in [23]. A small difference in the emission spectra under $\alpha$ particles and $\gamma$ rays excitation was observed. However, this effect could be due to different absorption of the light emitted by the localized source ($\alpha$ particels) or diffused one ($\gamma$ quanta).

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. 6, c (filled circles). Taking into account the reflection losses, the value of $\approx 10-15$ 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. 6, c. The crystals CWO–1 and CWO–3 show much better optical properties, namely the value of attenuation length is $\approx 50-70$ 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
Figure 6: (a) Emission spectra of CWO–5 crystal excited by $\gamma$ rays ($^{60}$Co) and $\alpha$ 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.

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 $\Omega$ (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 $\lambda$. 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.
2.4 Scintillation decay

2.4.1 Pulse shape for γ rays and α particles

Pulse shapes of CdWO₄ scintillators were studied as described in [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 (≈ 0.02 µs in comparison with ≈ 0.2 µs in [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 Ref. [42, 43]. Furthermore, single-electron counting method was applied to study the dependence of CdWO₄ scintillation signal for α particles and γ quanta on emission wavelength (see subsection 2.4.4).

To study pulse shape of scintillation decay for α particles, the CdWO₄ crystal 10 × 10 × 10 mm (CWO–6) was irradiated by α particles from collimated ²⁴¹Am source in the direction perpendicular to the (010) crystal plane. The dimensions of the collimator were ø0.75 × 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]. ⁶⁰Co was used as a source of γ quanta. Measurements were carried out at room temperature (23 ± 2)°C.

The shape of the light pulses produced by α particles and γ rays in the CdWO₄ 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 ²⁴¹Am) were summed. The first part of CdWO₄ 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, \( \tau_i \) – the decay constants for different light-emission components, and \( \tau_0 \) 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 \( \gamma \) rays and \( \alpha \) particles (see Table 3). Similar results have been obtained with the crystal CWO–7 studied both with the 20 MS/s and 125 MS/s digitizers.

![Figure 8: Decay of scintillation in CWO–6 crystal for \( \gamma \) rays and \( \alpha \) 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 \( \alpha \) particles with time decay of 0.14 \( \mu s \), 0.8 \( \mu s \), 4.1 \( \mu s \) and 14.1 \( \mu s \) are shown. Fitting functions for \( \alpha \) and \( \gamma \) pulses are shown by solid lines.](image)

### 2.4.2 Pulse-shape discrimination between \( \gamma \) rays and \( \alpha \) particles

The difference of the pulse shapes allows to discriminate \( \gamma(\beta) \) events from those induced by \( \alpha \) particles. We applied for this purpose the optimal filter method proposed in [14] and already applied to CdWO\(_4\) scintillators in [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 \( \mu s \) for 20 MS/s digitizer and 64 \( \mu s \) 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: \( P(t) = \{ f_\alpha(t) - f_\gamma(t) \} \).
Table 3: Decay time of CdWO$_4$ scintillators for $\gamma$ quanta and $\alpha$ particles measured by transient digitizers at room temperature. The decay constants and their relative intensities are denoted as $\tau_i$ and $A_i$, respectively.

| Type of irradiation | Decay constants (\mu s) and relative intensities |
|---------------------|-----------------------------------------------|
|                     | $\tau_1$ ($A_1$) | $\tau_2$ ($A_2$) | $\tau_3$ ($A_3$) | $\tau_4$ ($A_4$) |
| $\alpha$ particles  | 14.1 $\pm$ 0.3 (79.2 $\pm$ 2.0)$\%$ | 4.1 $\pm$ 0.6 (14.5 $\pm$ 1.2)$\%$ | 0.8 $\pm$ 0.2 (5.0 $\pm$ 0.6)$\%$ | 0.14 $\pm$ 0.07 (1.3 $\pm$ 0.4)$\%$ |
| $\gamma$ rays       | 14.5 $\pm$ 0.3 (88.7 $\pm$ 2.0)$\%$ | 4.6 $\pm$ 0.8 (8.7 $\pm$ 1.5)$\%$ | 0.8 $\pm$ 0.2 (2.1 $\pm$ 0.4)$\%$ | 0.15 $\pm$ 0.05 (0.5 $\pm$ 0.2)$\%$ |

\[ f_\gamma(t) / \{f_\alpha(t) + f_\gamma(t)\}, \] where $f_\alpha(t)$ and $f_\gamma(t)$ are the reference pulse shapes for $\alpha$ particles and $\gamma$ quanta.

Clear discrimination between $\alpha$ particles and $\gamma$ 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 $\alpha$ particles ($E_\alpha \approx 5.3$ MeV) and $\gamma$ quanta ($\approx 1.2$ MeV) are shown. As a measure of discrimination ability (factor of merit, $FOM$), the following expression can be used:

\[ FOM = |SI_\alpha - SI_\gamma| / \sqrt{\sigma_\alpha^2 + \sigma_\gamma^2}, \]

where $SI_\alpha$ and $SI_\gamma$ are mean SI values for $\alpha$ particles and $\gamma$ quanta distributions (which are well described by Gaussian functions, see Fig. 9), $\sigma_\alpha$ and $\sigma_\gamma$ 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 $\alpha$ particles and $\gamma$ 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 ($\lambda = 266$ 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. 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 $\approx 0.2$ $\mu$s, and sent to the input of a digital oscilloscope (HP TDS460). The
Figure 9: The shape indicator (see text) distributions measured by CWO–7 detector with \( \alpha \) particles \( (E_\alpha = 5.3 \text{ MeV}) \) and \( \gamma \) quanta \( (\approx 1.2 \text{ MeV}) \) using the 125 MS/s 12 bit transient digitizer. The distributions were fitted by Gaussian function (solid lines).

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 wavelength wavelengths are shown in Fig. 10. Three components of the scintillation decay with decay times and intensities \( \tau_1 \approx 15 \mu s \) (85\%), \( \tau_2 \approx 5 \mu s \) (11\%), and \( \tau_3 \approx 1 \mu s \) (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 \( \alpha \) particles and \( \gamma \) quanta at different wavelength of emission spectra

The pulse shape for \( \alpha \) particles and \( \gamma \) 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 \(-20^\circ\) 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
Figure 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.

Calibrator.

The pulse shapes of CdWO$_4$ scintillator for $\alpha$ particles ($^{241}$Am+$^{239}$Pu+$^{241}$Cm source) and $\gamma$ quanta ($^{137}$Cs) 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 $\alpha$ particles and $\gamma$ 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

| Type of irradiation | Decay constants ($\mu$s) and relative intensities |
|---------------------|-----------------------------------------------|
|                     | $\tau_1$ ($A_1$) | $\tau_2$ ($A_2$) | $\tau_3$ ($A_3$) | $\tau_4$ ($A_4$) |
| $\alpha$ particles  | $14.4 \pm 0.5$ | $4.5 \pm 0.7$ | $0.9 \pm 0.2$ | $0.15 \pm 0.03$ |
|                     | $(78.3 \pm 5.0)\%$ | $(16.0 \pm 4.0)\%$ | $(4.4 \pm 0.6)\%$ | $(1.3 \pm 0.4)\%$ |
| $\gamma$ rays       | $14.7 \pm 0.2$ | $4.2 \pm 0.4$ | $0.9 \pm 0.2$ | $0.11 \pm 0.04$ |
|                     | $(89.0 \pm 2.0)\%$ | $(8.4 \pm 1.0)\%$ | $(1.9 \pm 0.4)\%$ | $(0.7 \pm 0.2)\%$ |

Table 4: Decay time of CWO–7 scintillator for $\gamma$ quanta and $\alpha$ particles measured by single electron counting method at room temperature with 480 nm filter (see text). The decay constants and their relative intensities are denoted as $\tau_i$ and $A_i$, respectively.
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 $\alpha$ and $\gamma$ irradiation in the region of 420–590 nm.

2.4.5 Dependence of pulse shape on temperature

Temperature dependence of the pulse shape for $\gamma$ rays and $\alpha$ particles was checked in the range $0 - 25^\circ$ C. The CWO–4 crystal was optically connected to EMI9256KB PMT operating at −1000 volts. 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 $\gamma$ rays from $^{60}$Co source and $\alpha$ particles from $^{241}$Am source. Forms of signals for $\gamma$ rays and $\alpha$ particles have been obtained as a result of summation of several thousand 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 $\tau_1$ depends on the temperature as $-0.055(3) \mu$s/$^\circ$C for $\alpha$ particles and as $-0.048(3) \mu$s/$^\circ$C for $\gamma$ quanta. It should be noted, that the intensities of this component both for $\alpha$ and $\gamma$ signals remain constant: $(77.8 \pm 0.2)%$ for $\alpha$ particles and $(88.8 \pm 0.2)%$ for $\gamma$ quanta.

The temperature dependence of the averaged decay time is shown in Fig. 14, a. The averaged decay time decrease with temperature as $\approx -0.050(4) \mu$s/$^\circ$C for $\alpha$ particles.
and $\approx -0.048(7) \, \mu s/\degree C$ for $\gamma$ 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 $\tau_1$ decay component.

The factor of merit of pulse-shape discrimination between $\alpha$ particles and $\gamma$ quanta was calculated for the data accumulated in the temperature interval of $0 - 24\degree C$. The weight function (see subsection 2.4.2) was constructed by using pulse shapes for $\alpha$ particles and $\gamma$ quanta measured at room temperature. As one can see in Fig. 14, b, 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 $\alpha$ and $\gamma$ 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 $\gamma$ lines was obtained with large ($\varnothing 42 \times 39$ mm) CdWO$_4$ crystal scintillator. Small crystal ($10 \times 10 \times 10$ mm) showed an even better energy resolution: 6.8% and 3.4% for the 662 and 2615 keV $\gamma$ lines, respectively.
The absolute photon yield of CdWO₄ crystal scintillators was estimated to be $(30 - 41) \times 10^3$ photons per 1 MeV of energy deposit (under γ ray irradiation). This result was obtained by the analysis of the measurements of energy resolution. At least the lower border of this estimation is in agreement with the results of [26, 32]. In our opinion, more accurate measurements are necessary to determine the absolute light yield of CdWO₄.

Spectra of the low energy γ and X-ray lines (6 keV of ⁵⁵Fe, 18 keV of Neptunium L line and 60 keV γ quanta from ²⁴¹Am source) were measured, which demonstrates possibility to apply CdWO₄ crystal scintillators to search for double electron capture in ¹⁰⁶Cd. 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 [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 can not exclude that this effect is due to different absorption of scintillation light emitted under γ and α irradiation.

Transmissivity of CdWO₄ crystals was measured and considerable scattering of light was observed. This data indicates a presence of a substantial amount in our CdWO₄.
Figure 14: (a) Temperature dependence of the averaged scintillation decay time for $\gamma$ quanta ($\approx 1.2$ MeV) and $\alpha$ particles ($\approx 5.3$ 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.

crystal of optical heterogeneity whose sizes are comparable or exceed the scintillation light wavelength.

Four components of scintillation decay ($\tau_i \approx 0.1 - 0.2$, $\approx 1$, $\approx 4$ and $\approx 14 - 15$ $\mu$s) and their intensities under $\alpha$ particles and $\gamma$ quanta irradiation were measured with different CdWO$_4$ crystal scintillators by using different methods: transient digitizers with 20 MHz and 125 MHz sampling frequency as well as single electron counting method. The difference in the scintillation pulse shapes for $\alpha$ particles and $\gamma$ quanta is mainly due to difference in the intensities of the different decay components.

Clear discrimination between $\alpha$ particles and $\gamma$ 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 $\alpha$ particles and $\gamma$ quanta irradiation in the range 420–590 nm.

Temperature dependence of the decay constants and intensities of CdWO$_4$ pulse shape for $\gamma$ rays and $\alpha$ particles was investigated in the temperature range of $0 - 24^\circ$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 $\alpha$ particles and $\gamma$ quanta, while the intensities of this component both for $\alpha$ and $\gamma$ signals remain constant. The pulse-shape discrimination improved slightly with increasing of temperature.
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