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Pulse-shape discrimination with $CdWO_4$ crystal scintillators

T. Fazzini[a], P.G. Bizzeti[a,*], P.R. Maurenzig[a], C. Stramaccioni[a], F.A. Danevich[b,1], V.V. Kobychev[b,1], V.I. Tretyak[b,1], Yu.G. Zdesenko[b,1]

[a] *I.N.F.N. and Università di Firenze, Sezione di Firenze, L.go E. Fermi, 50125 Firenze, Italy*
[b] *Institute for Nuclear Research, 252028 Kiev, Ukraine*

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Abstract

The properties of light emission from a $CdWO_4$ crystal scintillator were investigated with different kinds of sources (α's, protons, cosmic muons and γ's). At least three components of scintillation signal were recognized with decay time of ≈ 1, 6 and 14 μs. By using the dependence of the relative amplitudes of these components on the nature of incoming radiation, the pulse-shape discrimination method based on the optimal digital filter was developed to process the scintillation pulses from the $CdWO_4$ crystal. The clear discrimination between γ-rays and α-particles was achieved, that permits the use of this technique in double-β-decay research and in other low-background measurements. © 1998 Elsevier Science B.V. All rights reserved.

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1. Introduction

Among inorganic scintillators, cadmium tungstate $CdWO_4$, has the attractive property of a very short attenuation length for γ-radiation, as well as excellent operational characteristics (chemical resistance, nonhygroscopicity, etc.) and has, therefore, been considered [1–10] together with bismuth germanate $Bi_4Ge_3O_{12}$, gadolinium orthosilicate Gd_2SiO_5, lead tungstate $PbWO_4$, as an alternative to NaI(Tl) for γ-detection. The important advantages of $CdWO_4$ crystal scintillators, apart from the high detection efficiency for X- and γ-rays, are the relatively large light yield (about 40% of NaI(Tl)), radiation resistance and very slight temperature dependence of the scintillation output [3–9]. The main disadvantage of the $CdWO_4$ scintillator is a quite long decay time (12–15 μs), which restricts the value of its maximum counting rate. However, this is not really important in some applications such as, for example, environmental measurements of radioactive contaminations. Also these crystals, due to their low level of radioactive contamination [11], can be utilized for the study of rare and forbidden processes in nuclear and particle physics. Indeed, these scintillators were used successfully in

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* Corresponding author. Tel.: 39 55 230751; fax: 39 55 229330.

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measurements of half-life and spectrum shape of the fourth-forbidden β-decay of ^{113}Cd ($T_{1/2} \approx 10^{16}$ y) [12] and in the search for possible α-decay of naturally occurring W isotopes [13]. Moreover, the experiment to study the double β-decay of ^{116}Cd was carried out with the help of $CdWO_4$ scintillators enriched with ^{116}Cd to 83%, which were developed for this research [9,14,15]. In spite of the small volumes of the crystals used (10–20 cm^3), one of the best results on the neutrinoless double-β-decay investigation was achieved [15,16]. In order to further advance this research and to improve the sensitivity of low-level measurements of radioactivities, $CdWO_4$ crystal scintillators of large volume (100–200 cm^3) with improved energy resolution have been developed [17] as a result of advances in the crystal growth, optimization of light collection and design of a special electronic unit for amplification and shaping of $CdWO_4$ signals. An energy resolution of 7–8% at 662 keV, comparable with NaI(Tl), has been obtained with small samples (10–20 cm^3), while for larger crystals (80–150 cm^3) the value was 10–12% for 662 keV γ-rays. With one such large crystal ($\approx$150 cm^3) a new limit of the half-life of the neutrinoless double β^+-decay of ^{106}Cd was obtained recently [18].

It is well known that an intrinsic limit to the sensitivity of the double-β-decay experiments is determined by radioactive contamination in the counter itself, in the photomultiplier, in the materials used for the detector mounting, and in the surroundings. In particular, α-decay from the families of natural radioactive isotopes – if contained in trace amounts within the counter – would produce pulses in an amplitude region which could overlap with the expected peak of neutrinoless ββ-decay. Actually, the energy available for the ββ-process is, in the case of ^{116}Cd, $E_{\beta\beta} = 2.8$ MeV, while the energy of α-particles from natural radioactivity ranges from 4 to 9 MeV. Also it is known, that scintillation efficiency of inorganic crystals depends on the local density of the energy released, and is usually smaller for highly ionizing particles than for electrons. Therefore, the possible use of pulse-shape discrimination (PSD) with $CdWO_4$ crystal scintillators can allow the reduction of background in these detectors and, as a result, promote ββ-decay research to a higher level.

The possibility of α/β pulse-shape discrimination with $CdWO_4$ crystals was demonstrated in Ref. [11] with the help of a comparison between the fast and total scintillation signal. But measured characteristics (energy threshold and discrimination factor) were not adequate for using this simple method in the experiment. Therefore, in the frame work of this discussion, the main goal of the present work was to develop the optimal PSD technique suitable for low-background measurements with $CdWO_4$ crystal scintillators.

An additional task which is connected with this goal is to investigate the properties of the light emission from $CdWO_4$ using different kinds of particles (α's, protons, minimum ionizing cosmic muons) and γ's to produce scintillation. Despite many studies of the scintillation properties of $CdWO_4$ [1–10,17], none of them (to our knowledge) has been devoted to the study of the possible dependence of the amplitude and shape of scintillation pulses on the mode of producing the crystal excitation.

As we will see in Sections 2–4, the scintillation efficiency decreases drastically from minimum ionizing particles to low-energy protons and α's, and also the time dependence of the light pulse changes enough to permit identification of the incoming particle by PSD technique. Discussion of the results and conclusions are presented in Section 5.

2. Experimental set-up

The crystal used for the measurement, supplied by INR Kiev [17], has cylindrical shape, 42 mm diameter and 25 mm height. It has been coupled to an EMI 9256B photomultiplier, the optical contact being ensured by a thin layer of silicone grease. Apart from a small window, through which low-energy protons and α's could enter, the crystal was surrounded and covered by a thin Teflon layer. The magnetic field on the photomultiplier was reduced by enclosing the photomultiplier and crystal in a μ-metal tube. Particular care has been taken to avoid any effect of charge loading that could alter the shape of the current pulse, with respect to that of the light signal.

The current pulse from the anode, integrated over an RC circuit having a time constant of 0.18 μs, was analyzed with the flash-ADC of a digital oscilloscope (Tektronix TDS460), operated at the sample frequency of 25 MS/s, and in more recent measurements, on an Analog Devices AD9022, 12 bit ADC, operated at the sample frequency of 20 MS/s. The time spectra, of up to 2500 channels, were transferred on-line to a personal computer, and stored in the mass memory for further analysis.

In a separate measurement, the same pulse provided the input for a charge-sensitive amplifier, having both the integration time and the fall time of the order of 50 μs, to investigate the amplitude spectra.

In addition to cosmic rays, γ- and X-rays from radioactive sources (^{57}Co, ^{60}Co, ^{137}Cs), α-particles from a source of mixed nuclides (^{239}Pu, ^{241}Am and ^{244}Cm) have been used. Protons of different energies, from 1.0 to 3.4 MeV, have been provided by the KN3000 accelerator of INFN, Firenze. The direct beam of the accelerator (of 20–30 nA) impinged on a thin Au foil, and part of the protons, scattered at about 45° through a series of collimators, could reach the $CdWO_4$ crystal. A thin window of kapton (of 3 mm diameter) separated the external counter from the vacuum system of the accelerator.

3. Results

3.1. Energy resolution and light yield

The energy resolution for γ-rays, with the time-shaping specified in the previous section, was equal to 8.3% for the 1.33 MeV γ-rays (Fig. 1). This value is comparable to the best reported so far [10,17], and much better than the older values reported, e.g., in Ref. [8]. Actually, the appearance of the present crystal was clean and transparent, and did not show any yellow pigmentation as the crystal described in Ref. [8]. In our case, the material appeared to be substantially transparent to its own radiation, at least over the dimensions of the crystal. In fact, the signal amplitude did not vary appreciably when the 120 keV γ-radiation of a ^{57}Co source entered the crystal from the side in a region adjacent to the photocathode or from the face opposite to the photomultiplier.

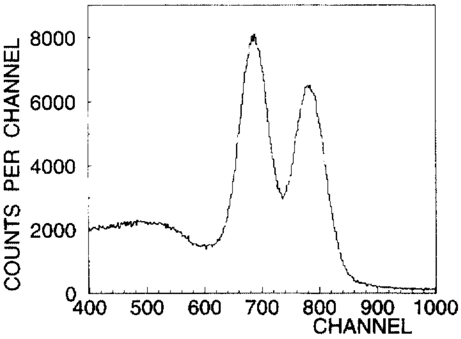

Fig. 1. Part of the spectrum of ^{60}Co γ-rays obtained with the $CdWO_4$ crystal. The energy resolution is 8.3%.

The attenuation length of 120 keV γ-rays in $CdWO_4$ is less than 1 mm. We can conclude that the light-collection efficiency does not depend appreciably on the scintillation point. For our purposes it is not relevant if this fact is a consequence of the small attenuation coefficient of the crystal to its own scintillation light, of the multiple reflections on the crystal surface making the average light path almost independent of the origin, or of both these reasons.

The comparison of pulse amplitudes produced by different sources of radiation gives the following results. After correction for energy loss in air and/or in windows, the light yield is reduced by almost a factor of 3 (or a factor of 10) when going from cosmic-rays and γ to low-energy protons (or α particles). The observed energy resolution turns out to be rather poor in our measurements with charged particles (15% for protons, 45% for alphas).

3.2. Pulse shape and decay time

Typical pulse shapes produced by α particles and cosmic-rays are depicted in Fig. 2A and B, respectively. Signals from cosmic-rays (and, possibly, gamma rays from the natural background) were selected in the energy window 2.2–3 MeV. Each line in Fig. 2 shows the average of a relatively large number of individual pulses. To obtain this result, raw data were first filtered by means of an interactive procedure to eliminate shapes whose amplitude exceeded ADC's range or clearly showing the presence of more than one pulse. Then,

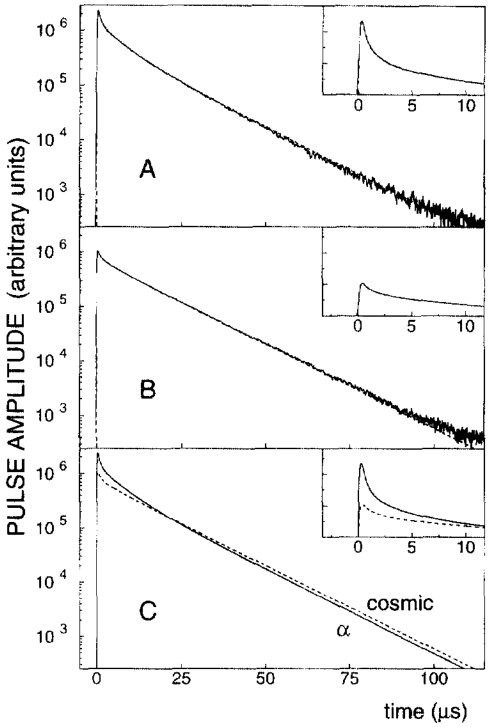

Fig. 2. Shape of the light pulses produced by minimum-ionizing particles of cosmic-rays (part B) and by low-energy α-particles (part A), compared with the analytic fit discussed in the text (dashed lines). Fitted shapes of α-pulses (full line) and of cosmic-ray pulses (dashed line), normalized to equal area, are compared in part C. In the insets, details of the very first part of each pulse are shown in a linear scale.

individual pulses were normalized to equal area and shifted, in order to make them to overlap exactly in time. A comparison of fitted pulse shapes for these two cases is presented in Fig. 2C that demonstrates the essential difference between them. This difference can be used to identify the radiation itself, as will be shown in the next section.

To provide an analytic description for each mode of excitation, the pulse shape resulting from the average of a large number of individual pulses has been fitted with a function corresponding to the sum of two or more exponentials, having an exponential rise time τ_0 and one or more decay times τ_k:

$$f(t) = \sum_k A_k \exp(-t/\tau_k) \quad (t > 0), \qquad (1)$$

with the condition $A_0 = -\sum_{k \geq 1} A_k$ in order to obtain $f(0) = 0$. The rise-time of the pulses resulted to be, in all cases, close to the integration time of the apparatus. None of the observed pulse shapes can be reproduced by fitting with one single decay time. Recent results by Kinloch et al. [10] have shown the presence of at least two components, with decay constants of about 1.1 and 14.5 µs. We could obtain a reasonable (although not excellent) fit in a limited region – between 2 and 50 µs from the beginning of the pulse – by assuming three components, two with time constants close to those of Ref. [10], and a third of about 6 µs. The latter component has been reported in some older works [8], which however referred to crystals with lower transparency to their own radiation.

In order to reproduce the shape of the pulses in the very first part, one more component with a much shorter time constant must be introduced. The exact value of the time constants, and particularly of the shortest one, are poorly determined by our data.[2] Values of 0.2, 0.9, 6 and 14 µs have been used to obtain the "fits" reported in Fig. 2. A rather accurate reproduction of the pulse shape in the entire region of interest ($0 < t < 50$ µs) is obtained in this way, and this is all we need for the present purpose. At later times, pulses show a long, weak tail, which is unimportant in the present context and has not been explicitly taken into account. Its presence, however, affects to some extent the estimate of the decay constants or at least of the longest one: its "best" value, in fact, is slightly smaller for α's than for minimum ionizing particles.

It should be stressed, however, that a substantial difference in the shape of scintillation pulses for α-particles and cosmic rays (or gammas), which is clearly visible from Fig. 2C, permits PSD with $CdWO_4$ crystal scintillators independent of any assumption about the number of time components.

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[2] In order to investigate the scintillation behavior of $CdWO_4$ crystals, more detailed measurements – and possibly resolved in wavelength – would be necessary. To this purpose, measurement with the UV light of a pulsed laser source would be appropriate.

4. Pulse-shape discrimination

The identification of particles entering a scintillation counter on the basis of the shape of the resulting light pulse is a well-known technique, which is widely used to reduce the γ-background in neutron counting with organic scintillators, and proved to work as well for a number of inorganic scintillators, such as CsI(Tl), NaI(Tl), BaF_2. The optimization of the signal processing for pulse shape discrimination is discussed, e.g. in Refs. [19,20]. Usually, the "optimal" procedure is simulated, as far as possible, by a proper analog filter. Here, a numerical filter has been used to process the scintillation signals, which – in fact – were already available in numerical form. First, the time origin of each signal was reconstructed with a digital "constant-fraction" technique, which is described in the Appendix. Then, two different digital filters are applied to the experimental signal, to obtain the "total charge" Q_t and the "pulse-shape discrimination" parameter Q_d:

$$Q_{t,d} = \sum_k P_{t,d}(t_k - t_0) f(t_k), \qquad (2)$$

and define the "shape parameter"

$$R = Q_d/Q_t. \qquad (3)$$

Good results are obtained with the following choice of the weight functions (corresponding to the "best linear filter" of Ref. [19] for constant variances): $P_t(t) = 1$, $P_d(t) = \bar{f}_\alpha(t) - \bar{f}_c(t)$ for $0 < t < t_{max} = 16\,\mu s$ and $P_t(t) = P_d(t) = 0$ elsewhere. Here $\bar{f}_\alpha(t)$ and $\bar{f}_c(t)$ are the average experimental pulse shapes (normalized to equal area) for alphas and cosmic muons (or gammas) which, for practical purposes, can be approximated by means of an analytic expression, like that described in Section 3.

Distributions of the values obtained for the shape parameter R are shown in Figs. 3 and 4. An excellent discrimination is obtained between α- and γ-pulses, for pulse-height corresponding to γ-ray (or electron) energies above 0.2 MeV. This is sufficient to identify (and, if necessary, eliminate) most of the alphas from radioactive contamination of the counter material.

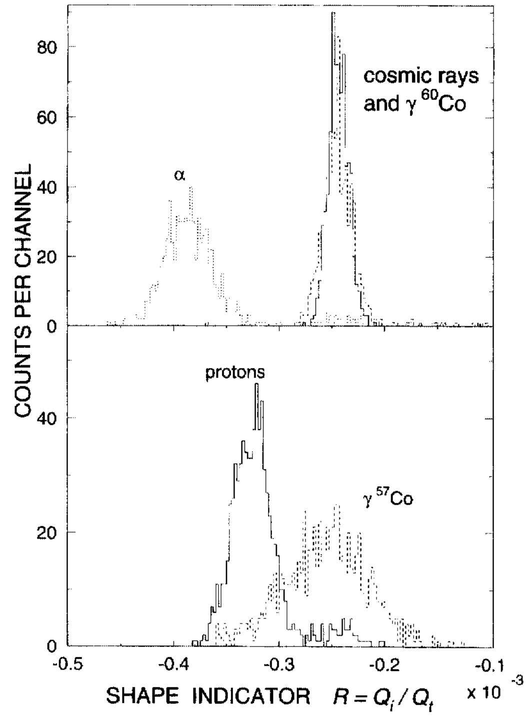

Fig. 3. Distribution of the "shape parameter" R for pulses produced by different modes of excitation. In part A, pulses from cosmic muons (full line) and ^{60}Co γ-rays (dashed line) are compared with those from α-particles of a radioactive source (dotted line). In part B, pulses from protons of 2.8–3.4 MeV (full line) and low-energy γ's from a ^{57}Co source (dashed line) are shown. The small satellite peak in the full-line distribution is due to low-energy γ's produced by inelastic scattering of the proton beam.

5. Discussion and conclusions

The spectral composition of the light emitted by $CdWO_4$ as described in Ref. [4] contains two separate components, one in the blue–green and one in the yellow region. The first one, corresponding to the shorter life-time, seems to be the intrinsic tungstate emission. The radiation of longer wavelength has been attributed [4] to defect centers. The decay time of the latter radiation was found to change with temperature in the region below 40 K, and to remain constant at higher temperature. However, the time constant reported in Refs. [4,8] is ≈ 20 μs, while in our case – as well as in recent Ref. [10] – the long-living component was measured to be

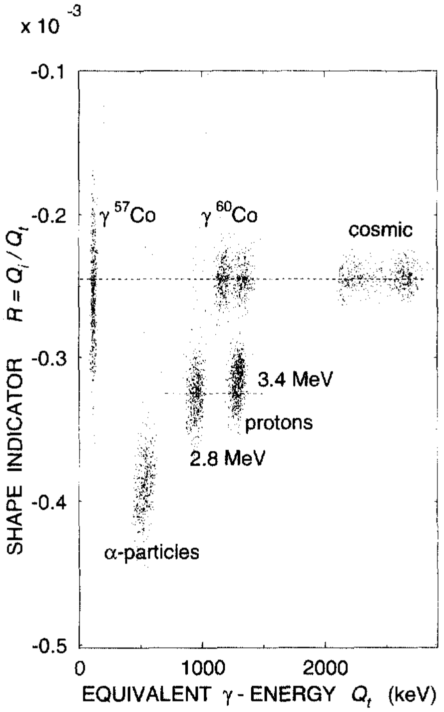

Fig. 4. Two-dimensional scatter plot for the pulses corresponding to different sources of radiation, with respect to the "total charge" parameter Q_t (horizontal axis) and to the "shape parameter" $R = Q_d/Q_t$ defined in the text. As – at least in the case of minimum ionizing particles or γ-rays – the parameter Q_d is proportional to the energy release, the "equivalent energy" E_γ is given in the horizontal scale.

definitely faster. The difference with respect to older values reported in the literature seems to be related to the different manufacturing procedure, possibly resulting in a different number and/or in different properties of the defect centers (and also, as a consequence, in a different color of the crystal).

In the present work the properties of the light emission from $CdWO_4$ crystal (decay time and shape of the light pulses) were investigated using different kinds of sources (alphas, protons, cosmic muons and gammas) to produce excitation of the scintillator.

At least three components of the scintillation signal were recognized with decay time of ≈ 1, 6 and 14 μs. The dependence of the relative amplitudes of these components on the mode of producing the crystal excitation was found to be sufficient for identification of the incoming radiation with the help of the PSD technique.

The pulse-shape discrimination method based on the optimal digital filter was developed and applied to process the scintillation pulses of $CdWO_4$ crystal. An excellent discrimination between γ-rays and α-particles was achieved, that permits the use of this technique in the experiment to study the double β-decay of ^{116}Cd [15,16] and in other low-background measurements.

In continuing our work with a different crystal, we have found a γ to α ratio of light yield of about 5, in agreement with Ref. [13]. The smaller light output for externally produced α-particles, observed with the previous crystal, has been found to be mainly due to the conditions of the crystal surface near the scintillation point. In all cases, the pulse shape discrimination remained equally good.

Appendix

The following procedure has been used to reconstruct the time origin of each signal with a digital "constant-fraction" technique. The numerical procedure corresponds to a time differentiation and a further smoothing with a Gaussian function (whose standard deviation σ must be optimized by trial and error):

$$\begin{aligned} F(t) &= \int (\mathrm{d}f(t')/\mathrm{d}t') G(t-t')\,\mathrm{d}t' \\ &= -\int f(t') G'(t-t')\,\mathrm{d}t', \end{aligned} \tag{A.1}$$

where $f(t)$ is the original time distribution of the signal, and

$$G(t-t') = \exp[-(t-t')^2/2\sigma^2], \tag{A.2}$$

$$\begin{aligned} G'(t-t' &= (\mathrm{d}/\mathrm{d}t)G(t-t') \\ &= -(t-t')\exp[-(t-t')^2/2\sigma^2]/\sigma^2. \end{aligned} \tag{A.3}$$

The first relative maximum of the function $F(t)$ (if not lower than $\frac{1}{4}$ of the absolute maximum) is evaluated by fitting over 4 adjacent points and taken as the time reference for the signal.

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