Investigation of β decay of ^{113}Cd

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β decay of ^{113}Cd was studied with the help of a low background $CdWO_4$ crystal scintillator (mass of 434 g) in an experiment at the Gran Sasso National Laboratories of the INFN for a period of 2758 h. The shape of the spectrum of the non-unique fourth-forbidden decay of ^{113}Cd was measured, and the half-life of ^{113}Cd was determined as $T_{1/2} = (8.04 \pm 0.05) \times 10^{15}$ yr.

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I. INTRODUCTION

^{113}Cd is one of only three nuclei which enables the investigation of four-fold forbidden β decays in a practical way, when rare transitions of this kind are not masked by much more rapid lower orders forbidden or allowed β decays (two other nuclides are ^{50}V and ^{115}In). The high order of forbiddeness is related to a large value of the half-life, in the range of 10^{16} yr, and radioactivity of ^{113}Cd was observed for the first time only in 1970 [1], after a number of unsuccessful attempts since 1940 (see references in Ref. [1]). In accordance with the last table of atomic masses, the energy release in the $^{113}\text{Cd} \rightarrow {}^{113}\text{In}$ decay is $Q_\beta = (320 \pm 3)$ keV [2].

In the experiment of Ref. [1], the cadmium foil with mass of $\simeq$0.6 g Cd enriched in ^{113}Cd to 96.38%[1] was measured (at the earth level) with a proportional counter over a period of 617 h. Comparison with measurements with a natural Cd sample leads to conclusions about the observation of the ^{113}Cd β decay with $T_{1/2} = (9.3 \pm 1.9) \times 10^{15}$ yr. The shape of the β spectrum was not studied in this experiment.

The first measurement of the ^{113}Cd β shape was reported in an experiment with a small (0.27 cm^3) CdTe semiconductor detector [4] (1988). The background to the ^{113}Cd β decay was not estimated, and the half-life value was conservatively derived as (4–12) $\times 10^{15}$ yr. The endpoint energy in the β decay was measured for the first time in direct observation as $Q_\beta = (320 \pm 10)$ keV. It should be noted that the measured β shape was quite different from those observed in subsequent experiments [5–7], probably because of big contributions from internal and external background sources.

A $CdWO_4$ crystal with mass of 58 g was used as a bolometer, at the temperature of 25 mK, in underground measurements at the Laboratori Nazionali del Gran Sasso [LNGS, 3600 m water equivalent (w.e.)] during 340 h [5] (1994). The good energy resolution of $\simeq$5 keV allowed the determination Q_β value precisely as 318.8 ± 1.4(stat.) $\pm$ 5(syst.) keV; the shape of the β spectrum was measured (see later Sec. IV C), and the half-life was given as [9.3 ± 0.5(stat.) $\pm$ 1(syst.)] $\times 10^{15}$ yr. Nevertheless, the background in the ^{113}Cd β decay region was quite large, with signal-to-background (S/B) ratio near 2/1.

In 1996, a $CdWO_4$ crystal with mass of 454 g was used as a scintillator in the experiment in the Solotvina Underground Laboratory (1000 m w.e.) [6]. With $S/B = 50$, the half-life was precisely determined as $(7.7 \pm 0.3) \times 10^{15}$ yr, the shape of the spectrum was measured, and the energy release was estimated as $Q_\beta = [337.4 \pm 0.3(\text{stat.}) \pm 22(\text{syst.})]$ keV, the last uncertainty being related to the energy resolution of the scintillator ($\sigma \simeq 22$ keV at the energy of 337 keV).

In a recent (2005) experiment at the LNGS, three semiconductor detectors $Cd_{0.9}Zn_{0.1}Te$ with volume near 1.1 cm^3 each were measured over a period of 4781 h to derive the ^{113}Cd half-life $T_{1/2} = [8.2 \pm 0.2(\text{stat})^{+0.2}_{-1.0}(\text{syst})] \times 10^{15}$ yr [7]. Due to variations of the Zn amounts in the $Cd_{0.9}Zn_{0.1}Te$ material, the uncertainty in number of ^{113}Cd nuclei was estimated as 1.8%. The Q_β value was not derived independently; however, the end of the spectrum was in agreement with the accepted value of 320 keV. While in this experiment the spectrum shape could be measured, the high energy threshold (near 100 keV) did not allow one to do this precisely; and for the $T_{1/2}$ derivation, the spectrum shape of Ref. [6] was used.

The summary of all up-to-date experiments is given in Table I. The decay scheme of ^{113}Cd is shown in Fig. 1.

The aim of the present study is to use a low background $CdWO_4$ crystal scintillator to investigate the β decay of ^{113}Cd (half-life value and spectrum shape) with better precision than in previous experiments (4–12% uncertainty for $T_{1/2}$ in the recent ones). To achieve this, in addition to the low background, low energy threshold, and good spectrometric characteristics of the scintillator, the number of ^{113}Cd nuclei should also be accurately determined because the recommended IUPAC value of ^{113}Cd abundance 12.22(12)% [3] already introduces a 1% uncertainty. The $CdWO_4$ crystal exploited in the experiment

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[1]The natural abundance of ^{113}Cd is 12.22% [3].

TABLE I. Summary of experiments on ^{113}Cd β decay. $N(^{113}\mathrm{Cd})$ is the number of ^{113}Cd nuclei available for the investigation; E_{th} is the energy threshold for the β spectrum measurement; FWHM is the energy resolution of the detector, and S/B is the signal to background ratio.

Experimental method	$N(^{113}\mathrm{Cd})$	E_{th} (keV)	FWHM (keV)	S/B ratio	Half-life (10^{15} yr)	Year Ref.
PC[a] + ^{113}Cd foil 0.56 g (617 h, earth level)	3.0×10^{21}	–	–	$\simeq 1/20$	9.3 ± 1.9	1970 [1]
CdTe[b] 0.27 cm^3 (433 h, 3 m w.e.)	5.0×10^{20}	$\simeq 30$	$\simeq 50$[c]	–	4–12	1988 [4]
$CdWO_4$[d] 58 g (340 h, 3600 m w.e.)	1.2×10^{22}	$\simeq 20$	$\simeq 5$[c]	$\simeq 2$	9.3 ± 1.1	1994 [5]
$CdWO_4$[e] 454 g (433 h, 1000 m w.e.)	9.3×10^{22}	44	$\simeq 49$[f]	50	7.7 ± 0.3	1996 [6]
CdZnTe[b] 3×1.1 cm^3 (4781 h, 3600 m w.e.)	6.1×10^{21}	$\simeq 100$	$\simeq 43$[f]	$\simeq 8$	$8.2^{+0.3}_{-1.0}$	2005 [7]
$CdWO_4$[e] 434 g (2554+2758 h, 3600 m w.e.)	8.9×10^{22}	28	$\simeq 47$[f]	56	8.04 ± 0.05	Present work

[a]Proportional counter.
[b]Semiconductor detector.
[c]Independent of energy.
[d]Low-temperature bolometer.
[e]Crystal scintillator.
[f]At energy 320 keV.

[6] was used in the present measurements, after storage during the last 10 yr in the Solotvina Laboratory at a depth of 1000 m w.e. Thus, it was not exposed to cosmic rays during this period, and new cosmogenic activities were not induced, while the old ones (if any) had time to decrease or disappear.

The main properties of the $CdWO_4$ scintillator are (i) density equal to 7.9 g/cm^3, (ii) light yield $\approx$30–40% of that of NaI(Tl), (iii) refractive index equal to 2.2–2.3, (iv) emission maximum at 480 nm, and (v) effective average decay time of 13 μs. The material is nonhygroscopic and chemically inert; the melting point is at 1271 °C. The radiopurity of cadmium tungstate crystals has been investigated in several low background experiments (see Refs. [9–12] and references therein).

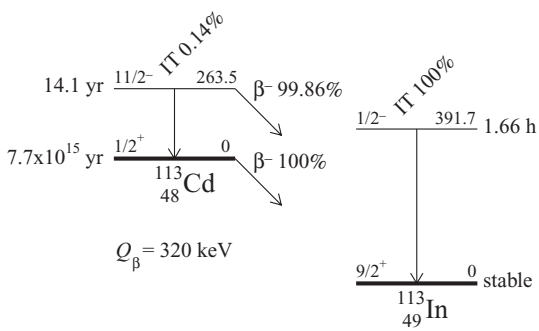

FIG. 1. Nuclear decay scheme of the ^{113}Cd-^{113}In doublet in accordance with Ref. [8].

II. MEASUREMENTS

A. Detector

The low background $CdWO_4$ crystal scintillator 40 mm in diameter by 43 mm in length (mass of 433.61 g) was produced from the crystal applied in the experiment carried out in the Solotvina Underground Laboratory [6]. After the measurements in 1995, the crystal was stored in the Solotvina Underground Laboratory for 10 yr at a depth of 1000 m w.e. In August 2005, the crystal was transported in a lead container (with walls $\approx$12 cm thick) by surface and immediately placed underground in the Gran Sasso National Laboratories of the I.N.F.N. (3600 m w.e.) to avoid cosmogenic activation. The crystal was washed by ultrapure nitric acid and vacuum packed to prevent contact with radon from air. The measurements comprising the experiment were started in January 2006.

The $CdWO_4$ crystal was fixed inside a cavity $\oslash 47 \times 59$ mm in the central part of a polystyrene light guide, 66 mm in diameter and 312 mm in length. The cavity was filled with high-purity silicon oil. The light guide was optically connected on opposite sides by a Dow Corning Q2-3067 optical couplant to two low radioactive EMI9265–B53/FL, 3-in.-diameter photomultipliers (PMTs). The light guide was wrapped by a polytetrafluoroethylene (PTFE) reflection tape.

The detector was installed deep underground in the low background DAMA/R&D setup at the Gran Sasso National Laboratory of the I.N.F.N. The detector was surrounded by Cu bricks and sealed in a low radioactive airtight Cu box and continuously flushed with high-purity nitrogen gas (stored deeply underground for a long time) to avoid the presence of residual environmental radon. The Cu box was surrounded by a passive shield made of high-purity Cu, 10 cm thick, 15 cm

of low radioactive lead, 1.5 mm of cadmium, and 4–10 cm of polyethylene/paraffin to reduce the external background. The shield was contained inside a Plexiglas box, also continuously flushed by high-purity nitrogen gas.

An event-by-event data acquisition system recorded amplitude and arrival time of events. Moreover, the sum of the signals from the PMTs was also recorded by a 1 GSample/s 8 bit DC270 transient digitizer by Acqiris over a time window of 100 μs. Taking into account the slow kinetics of the $CdWO_4$ scintillation decay ($\approx$13 μs), a sampling frequency of 20 MSample/s was used during the experiment.

B. Low background measurements

The experiment was carried out in the underground Gran Sasso National Laboratories of the INFN at a depth of 3600 m w.e.

First, the data were accumulated over 2554 h in the energy interval up to $\approx$1.7 MeV (run 1). Unfortunately, the energy resolution was rather poor during this run (full width at half maximum, FWHM = 13.3% for 1461 keV γ line of ^{40}K present in the background spectrum) because of some degradation of optical contact between the light guide used in the present experiment and one of the PMTs. Nevertheless, the data of run 1 were used to estimate the radioactive contamination of the $CdWO_4$ crystal. Then the experiment was carried out for 2758 h with the upper energy threshold $\approx$0.6 MeV to investigate precisely the β spectrum of ^{113}Cd (run 2).

The energy scale and resolution of the $CdWO_4$ detector for γ quanta were measured with ^{22}Na, ^{133}Ba, ^{137}Cs, ^{228}Th, and ^{241}Am sources (see Fig. 2). The energy dependence of the energy resolution can be fitted by the function $\mathrm{FWHM}_\gamma(\mathrm{keV}) = \sqrt{6.8(4)E_\gamma}$, where E_γ is the energy of γ quanta in keV. The energy scale was reasonably stable with deviations in the range of 2–3%.

The energy spectrum accumulated in the low background setup with the $CdWO_4$ detector during run 2 is presented in Fig. 3. The counting rate in the spectrum below the energy of $\approx$380 keV is mainly provided by the β decay of ^{113}Cd.

Because of the features and light response of the used $CdWO_4$ detector, PMT noise could contribute up to a few tens keV to the measured energy spectrum (see later). To estimate the shape of the PMT noise spectrum, a "blank" run was carried out at the end of the experiment. For this purpose, the $CdWO_4$ crystal was removed from the used light guide, the setup was closed, and measurements were carried out under the same conditions. More than 1 million noise events have been recorded.

C. Mass spectrometric measurements of the $CdWO_4$ crystal

1. *Isotopic abundance of* ^{113}Cd

Precise mass spectrometric measurements of the isotopic abundance of the $CdWO_4$ crystal were performed in the Department of Applied Physics of Curtin University (Perth, Australia) to determine the number of ^{113}Cd nuclei in the

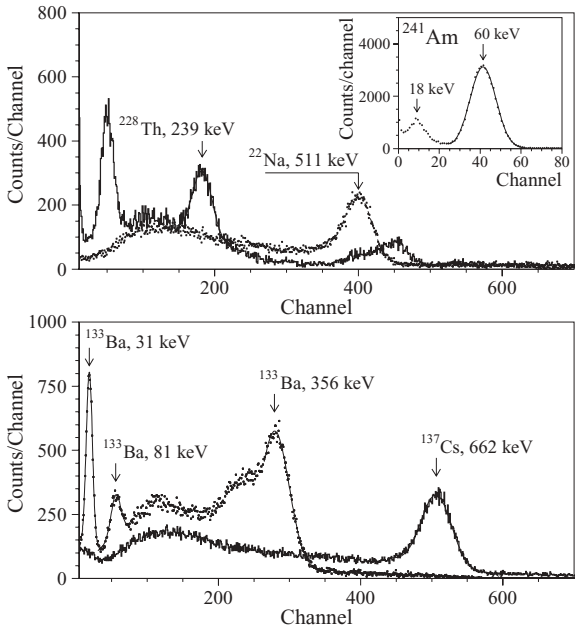

FIG. 2. Energy spectra accumulated by the used $CdWO_4$ detector with ^{22}Na, ^{228}Th, ^{241}Am (top figure), ^{133}Ba, and ^{137}Cs γ sources (bottom figure).

crystal. For accurate mass spectrometric analysis, a sample of the $CdWO_4$ crystal was taken partially into solution and purified by ion exchange chemistry to successfully extract Cd from the rest of the material. The sample was then loaded onto a single Re filament assembly and prepared for mass spectrometric analysis. The sample was measured by thermal ionization mass spectrometry using the Daly and the Faraday

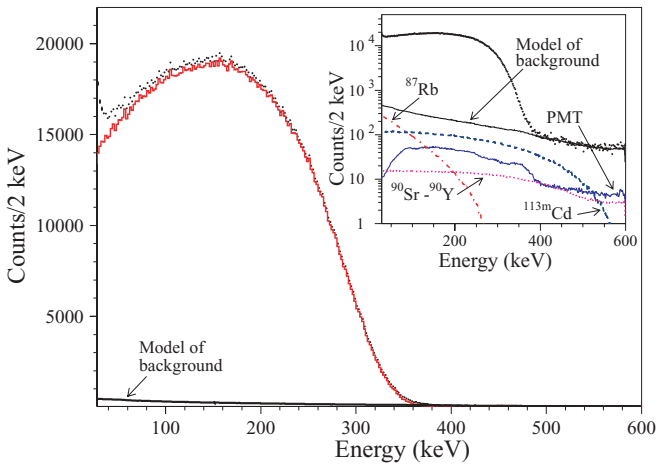

FIG. 3. (Color online) Raw energy spectrum of the $CdWO_4$ scintillator measured over 2758 h in the low background setup is shown by dots. The energy spectrum, obtained after the PMT noise rejection and the correction for related efficiency and the subtraction of the background, is shown by histogram. Inset: Raw spectrum together with the model of the background and its main components: β spectra of ^{87}Rb, ^{113m}Cd, and ^{90}Sr-^{90}Y, and the contribution from the external γ quanta from PMTs in these experimental conditions.

cup collector system. The $CdWO_4$ samples were compared to the International Reference Material BAM (Bundesanstalt für Materialforschung und-prüfung)–spectroscopically pure Cd laboratory standard, in order to examine the difference of the isotopic composition of the Cd in the $CdWO_4$ material with that of the Cd standard. The chemical and instrumental fractionations that may be introduced through ion exchange as well as by the thermal ionization processes of the mass spectrometer were examined and corrected by using both conventional means and a double spiking methodology. The abundance of ^{113}Cd in the $CdWO_4$ crystal was determined as 12.22(0.04)% (with the relative uncertainty at the 2σ level). The accuracy of this measurement corresponds to the "best measurement from a single terrestial source" [13] and is three times better than the one recommended by IUPAC [3,14]. So, the number of ^{113}Cd nuclei in the crystal used in the present experiment can be calculated on the basis of mass spectrometry measurements as $N = (8.858 \pm 0.014) \times 10^{22}$.

2. *Impurities in the* $CdWO_4$ *crystal*

To estimate the presence of naturally occurring β and α active isotopes, the $CdWO_4$ crystal was measured with the help of an inductively coupled plasma-mass spectrometry analysis (ICP-MS, Agilent Technologies model 7500a) at LNGS. A sample of the $CdWO_4$ crystal was reduced to powder by mechanical treatment inside a cleaned polyethylene bag to avoid possible external contamination. Two portions of the sample were etched in a microwave-assisted acid digestion technique (Method EPA 3052) using nitric acid and nitric-hydrofluoric acid mixtures. The results for both procedures were just a partial sample dissolution. The solutions obtained after centrifugation have been analyzed by ICP-MS.

The concentrations were measured by a semiquantitative technique: the instrumentation was calibrated with a single standard using a solution containing four elements to cover the entire mass range. The results of the measurements are presented in Table II. The errors of the values are at the level of 20–30%. Moreover, the errors for some elements (in particular, for In and Th) could be higher. For instance, inaccuracy in the In contamination was mainly due to the fact that a few weeks before, a sample of tin had been measured in the mass spectrometer, and the device was still slightly contaminated by Sn. At the same time, the most abundant isotope of In is 115, which is the same as a minor isotope of Sn. The error for Th (atomic mass 232) could be due to interference with tungsten oxide, $^{184}W^{16}O_3$. The values in Table II are the average of the measurements on the two solutions which were in good agreement.

TABLE II. Contamination of the $CdWO_4$ crystal scintillator measured by ICP-MS analysis.

Element	Measured atomic mass	Concentration of element (ppb)	Possible interference
V	51	3.8	
Ni	60	250	
Rb	85	3.2	
Ru	101	$\leqslant$3	
In	115	8.5	^{115}Sn
Sb	121	5.9	
Ce	140	$\leqslant$2	
Sm	147	$\leqslant$3	
Pt	195	3	
Pb	208	340	
Th	232	0.7	$^{184}W^{16}O_3$
U	238	$\leqslant$0.6	

III. DATA ANALYSIS

A. PMT noise rejection

The time characteristics of $CdWO_4$ scintillators under γ and α irradiation were investigated in Refs. [15–17]. Four components with decay constants $\tau_i \approx 0.1$–$0.2, \approx 1, \approx 4$, and $\approx$14–15 μs and different amplitudes for γ rays and α particles were observed.

The time characteristics of PMT noise in the present experimental condition were studied in a special blank run as mentioned in Sec. II B. The noise signals have the simple form of very fast single pulses; casual presence of more than a single PMT noise signal could occur in the present experiment over a very long 100 μs time window (recall the large time decay of $CdWO_4$ scintillation pulses and its light response).

To effectively discriminate the PMT noise, as in a previous experiment [18], two approaches were applied: the optimal filter technique proposed in Ref. [19] and developed in Ref. [15] for $CdWO_4$ crystal scintillators, and the mean time method [20]. In both cases, the pulse shape was investigated from the origin of the pulse and up to 50 μs. The pulse-shape discrimination technique to reject PMT noise was checked with the help of the calibration data. The energy dependence of efficiency to select scintillation signals was estimated as well; the efficiency is about 0.8 at 30 keV and about 1 above roughly 50 keV (consider the features and the light response of the $CdWO_4$ detector used).

In Fig. 4, the 28–100 keV energy region of the energy spectrum measured by the $CdWO_4$ scintillator during a period of 2758 h in the low background setup is depicted. It shows the spectrum (i) before PMT noise rejection, (ii) after PMT noise rejection corrected for related efficiency, and (iii) after background subtraction. It also shows the fit of the spectrum by a polynomial function (see also later).

B. Radioactive contamination of the $CdWO_4$ detector

The knowledge of the $CdWO_4$ crystal radioactive contamination is necessary to reconstruct the background in the energy interval of the β spectrum of ^{113}Cd. The time-amplitude analysis, the pulse-shape discrimination between $\beta(\gamma)$ and α particles, the simulation of the measured energy spectrum, and the data of the mass spectrometry measurements by

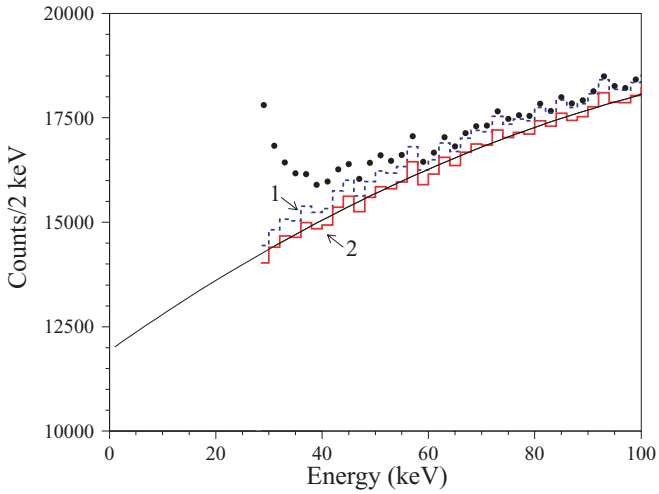

FIG. 4. (Color online) Low energy part (28–100 keV) of the energy spectrum of the $CdWO_4$ scintillator during a period of 2758 h in the low background setup. The spectrum is shown (i) before PMT noise rejection (dots), (ii) after PMT noise rejection corrected for related efficiency (histogram 1), and (iii) after background subtraction (histogram 2). The solid line represents the fit of the spectrum (2) by a polynomial function (see also later).

ICP-MS were used to estimate the radioactive contamination of the $CdWO_4$ detector.

1. *Estimation of* ^{228}Th, ^{227}Ac, *and* ^{226}Ra *activity by time-amplitude analysis*

The activities of ^{228}Th (^{232}Th family), ^{227}Ac (^{235}U), and ^{226}Ra (^{238}U) in the $CdWO_4$ crystal were determined with the help of the time-amplitude method using the data of run 1. The arrival time and energy of each event were used for the selection of fast decay chains in the ^{232}Th, ^{235}U, and ^{238}U families[2].

To estimate the activity of ^{228}Th, the following sequence of α decays was searched for and observed: ^{220}Rn ($Q_\alpha = 6.405$ MeV) $\rightarrow$ ^{216}Po ($Q_\alpha = 6.907$ MeV, $T_{1/2} = 0.145$ s) $\rightarrow$ ^{212}Pb. In $CdWO_4$ scintillators, α particles produce a substantially lower scintillation in comparison with γ quanta (β particles). Numerically this quenching can be expressed through the α/β ratio. We have used the energy dependence of the α/β ratio measured in Ref. [16] for $CdWO_4$ crystal scintillators: $\alpha/\beta = 0.083(9) + 0.0168(13)E_\alpha$, where E_α is the energy of the α particles in MeV. Thus, all events within 0.75–1.75 MeV were used as triggers, while a time interval 0.01–1 s (90.4% of ^{216}Po decays) and the same energy window were set for the second events. The obtained α peaks as well as the distribution of the time intervals between the first and second events are in good agreement with those expected for α particles of the ^{220}Rn $\rightarrow$ ^{216}Po $\rightarrow$ ^{212}Pb chain. On this basis the activity of ^{228}Th in the $CdWO_4$ crystal was calculated as 0.008(4) mBq/kg.

[2]The technique of the time-amplitude analysis is described in detail in Refs. [21–23].

TABLE III. Radioactive contamination of $CdWO_4$ scintillator determined by different methods. Activities are in mBq/kg.

Chain	Nuclide	Method of estimation and activity			
		t-A[a]	PSD[b]	Fit of background spectrum	ICP-MS[c]
^{232}Th	^{232}Th		$\leqslant$0.026	$\leqslant$0.04	3.1[d]
	^{228}Th	0.008(4)	$\leqslant$0.014	$\leqslant$0.018	
^{235}U	^{227}Ac	0.014(9)			
^{238}U	^{238}U		$\leqslant$0.045	$\leqslant$0.1	
	^{230}Th		$\leqslant$0.18		
	^{226}Ra	$\leqslant$0.04	$\leqslant$0.018	$\leqslant$0.03	
	^{210}Po		$\leqslant$0.063	$\leqslant$0.12	
Total α activity			0.26(4)		
	^{40}K			$\leqslant$5	
	^{60}Co			$\leqslant$0.4	
	^{87}Rb				3.1
	^{90}Sr-^{90}Y			$\leqslant$1	
	^{113}Cd			558(4)	
	^{113m}Cd			$\leqslant$3.4	
	^{137}Cs			$\leqslant$0.3	
	^{147}Sm		$\leqslant$0.01		

[a]Time-amplitude analysis (see Sec. III B1).
[b]Pulse-shape discrimination (see Sec. III B2).
[c]Inductively coupled plasma - mass spectrometry (see Sec. II C2).
[d]See remark in Table II about possible interference with $^{184}W^{16}O_3$ molecule.

The same technique was applied to the sequence of α decays from the ^{235}U family: ^{219}Rn ($Q_\alpha = 6.946$ MeV) $\rightarrow$ ^{215}Po ($Q_\alpha = 7.526$ MeV, $T_{1/2} = 1.78$ ms) $\rightarrow$ ^{211}Pb. The obtained α peaks correspond to the activity of 0.014(9) mBq/kg for the ^{227}Ac impurity in the crystal.

The following sequence of decays was used to estimate ^{226}Ra contamination in the detector: ^{214}Bi ($Q_\beta = 3.272$ MeV) $\rightarrow$ ^{214}Po ($Q_\alpha = 7.833$ MeV, $T_{1/2} = 164$ μs) $\rightarrow$ ^{210}Pb. The sensitivity of the method for this chain is rather low in our case because of the comparatively large minimal time between signals that the data acquisition system was able to record separately (using the calibration data, this time interval was determined as 3.16 ms). Only a couple of events (which can be ascribed to this fast sequence) were selected, and the limit on activity of ^{226}Ra in the $CdWO_4$ crystal was set as $\leqslant$0.04 mBq/kg.

All the activities obtained by the time-amplitude analysis are presented in Table III.

2. *Pulse-shape discrimination between* $\beta(\gamma)$ *and* α *particles*

As was demonstrated in Refs. [15–17], the difference of the pulse shapes in the $CdWO_4$ scintillator allows discrimination of $\gamma(\beta)$ events from those induced by α particles. The optimal filter method was applied for this purpose. 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, respectively. As an example, the

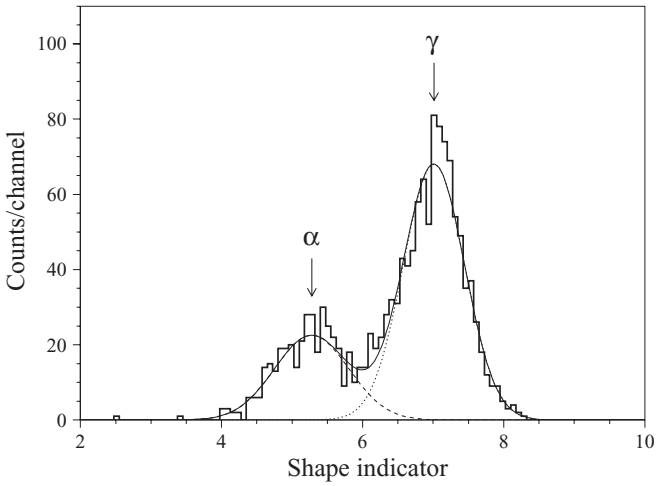

FIG. 5. Distributions of the shape indicator (see text) for pulses produced by background γ quanta (β particles) and α particles in $CdWO_4$ scintillation detector. The distributions were fitted by Gaussian functions (solid lines).

distribution of the shape indicator (see Refs. [15–17,19]) for $\beta(\gamma)$ and α events in the energy interval 700–800 keV (data of the run 1) is shown in Fig. 5.

The energy spectra of $\gamma(\beta)$ and α events selected with the help of the pulse-shape discrimination from data of run 1 are shown in Fig. 6. As was demonstrated in Ref. [16], the energy resolution for α particles is worse than that for γ quanta because of the dependence of the α/β ratio on the direction of α particles relative to the $CdWO_4$ crystal axes. The energy resolution FWHM_α (keV) $= 33 + 0.247E_\alpha^\gamma$ (here E_α^γ is the energy of α particles in γ scale expressed in keV) obtained in Ref. [16] was used in the present analysis. Fitting the spectrum

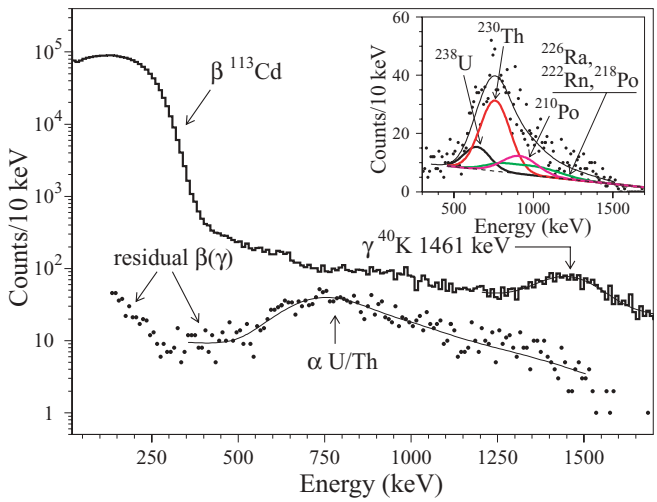

FIG. 6. (Color online) Energy spectrum of β particles (γ quanta, solid histogram) and α particles (dots) selected by the pulse-shape discrimination from raw data measured with the $CdWO_4$ scintillator during 2554 h (run 1) in the low background setup. In the inset, the α spectrum is depicted together with the model, which includes α decays from ^{238}U family. The total α activity in the $CdWO_4$ crystal is 0.26 mBq/kg.

by simulated α peaks from U/Th, we obtain activities of the U/Th daughters in the crystal (equilibrium of U/Th chains was supposed to be broken). However, because of the poor energy resolution for α particles, we can only give limits on the activities of U/Th daughters in the crystal (see Table III). The total internal U/Th α activity in the detector is 0.26(4) mBq/kg.

There could be a possible contamination of the crystal by ^{147}Sm ($E_\alpha = 2233$ keV). An α peak of ^{147}Sm is expected at the energy of 270 keV with FWHM = 100 keV. The ^{147}Sm contamination was checked by the analysis of the α spectrum in the energy interval 220–500 keV. The simple model, including the expected peak and polynomial function of the second degree, was taken to describe the background. The fit only gives a limit $\leqslant 0.01$ mBq/kg for the activity of ^{147}Sm in the crystal.

3. *Simulation of $\beta(\gamma)$ background*

There are several β active radionuclides which could produce background in the energy region of the ^{113}Cd β spectrum. The most significant of them are listed in Table III. Radioactive contamination of the PMTs could also contribute to the background. The distributions of the possible background components were simulated with the help of the GEANT4 package [24]. The initial kinematics of the particles emitted in the decay of nuclei was given by an event generator DECAY0 [25]. To estimate the contribution of the β active isotopes and of the γ rays from the PMTs, the measured spectrum of the $CdWO_4$ detector (Fig. 3) was fitted by the model built from the simulated distributions. Activities of U/Th daughters in the crystal were bounded taking into account the results of the pulse-shape and time-amplitude analyses. The activities of ^{40}K, ^{232}Th, and ^{238}U inside the PMTs were taken from Ref. [26]. There are no clear peculiarities in the spectrum which could be ascribed to the internal trace contamination by radioactive nuclides. Therefore we can obtain only limits on the activities of ^{40}K, ^{60}Co, ^{90}Sr-^{90}Y, ^{113m}Cd, ^{137}Cs, and U/Th daughters. The result of fit in the energy region 388–600 keV (χ^2/n.d.f. $= 115/92 = 1.25$) and the main components of the background are shown in Fig. 3.

The summary of the measured radioactive contamination of the $CdWO_4$ crystal scintillator (or limits on their activities) is given in Table III.

We have also checked possible contributions of cosmogenic long-lived (with $T_{1/2}$ in the range of years) pure β active radionuclides which could be created in the crystal due to interaction with cosmic rays during its stay on the earth surface: ^{14}C ($Q_\beta = 157$ keV, $T_{1/2} = 5730$ yr), ^{32}Si (224 keV, 172 yr), ^{39}Ar (565 keV, 269 yr), ^{135}Cs (269 keV, 2.3×10^6 yr), and others. The activities were calculated with the COSMO code [27]. We have found that these nuclides give negligible contribution to the area of investigated β spectrum of ^{113}Cd because of the relatively low probability of their production. As for the short-lived cosmogenic radionuclides, their activities (1) were not evident in our previous experiment in 1995 [6] and (2) should be reduced further during 10 yr of

storage of the crystal underground in the Solotvina Laboratory (1000 m w.e.).

Finally, the contribution from the $2\nu2\beta$ decay of ^{116}Cd ($Q_{2\beta} = 2809$ keV, $T_{1/2} = 2.9^{+0.4}_{-0.3} \times 10^{19}$ yr [12]) is negligible: with an activity of 95 μBq/kg, it gives $\approx$30 events in the energy interval 0–380 keV compared to 2.4×10^6 events from ^{113}Cd β decay.

IV. RESULTS AND DISCUSSION

A. Half-life of ^{113}Cd

Below we determine the half-life of ^{113}Cd in the most model-independent way. To calculate the $T_{1/2}$ value, the behavior of the spectrum at low energies has to be determined. It is worthwhile to note that the main part of the error in $T_{1/2}$ and in the spectral shape of the experiment of Ref. [6] was related with the uncertainty of the shape of β spectrum below the energy threshold of 44 keV.

The model of the background in the energy interval of the β spectrum was built as described in Sec. III B3. The activity of ^{87}Rb was taken to be 3.1 mBq/kg using the results of the ICP-MS analysis. The model of background and its main components are shown in Fig. 3. The background in the present experiment is only $\approx$1.8% of the β spectrum of ^{113}Cd. The effect to background ratio thus is 56/1, which is the best value among all the experiments that have studied the β decay of ^{113}Cd.

The energy spectrum, after the PMT noise rejection and the correction for related efficiency, and after subtraction of the background, is shown in Fig. 3 by histogram; the low energy part of the spectrum is also presented in Fig. 4. At low energies, the shape of the β spectrum was approximated by a polynomial function of the fourth degree. With this aim, the energy spectrum was fitted in the energy intervals from (20–40) to 260 keV (the lower bound of the interval was changed with the step of 2 keV). The best fit (χ^2/n.d.f. = 137/111 = 1.23) was achieved in the energy interval 30–260 keV. The result of the fit is presented in Fig. 4. Then, the number of events in the β spectrum was calculated as the area of the polynomial function below the energy of 30 keV plus the number of events in the experimental spectrum in the energy interval 30–390 keV. It gives the number of events in the β spectrum of ^{113}Cd: $S = [2403374 \pm 1490(\text{stat.})^{+13460}_{-12860}(\text{syst.})]$ counts. Therefore, the β activity of ^{113}Cd in the $CdWO_4$ crystal is 558(4) mBq/kg.

The half-life of ^{113}Cd can be calculated according to the formula: $T_{1/2} = \ln 2 \eta N(t/S)$, where N is number of ^{113}Cd nuclei in the crystal, η is efficiency of registration of the β decay of ^{113}Cd, and t is the time of measurements.

The efficiency η of registration of the β decay of ^{113}Cd by the $CdWO_4$ detector was calculated with the help of the GEANT4 code. One million decays were simulated with vertexes uniformly distributed over the crystal volume. Efficiency of total absorption of β particles was calculated as 99.97%. It leads to the half-life of ^{113}Cd: $T_{1/2} = [8.037 \pm 0.005(\text{stat.})^{+0.045}_{-0.043}(\text{syst.})] \times 10^{15}$ yr.

The statistical error in the present experiment is much lower than the systematic error (the main sources of the systematic error and their contributions to the value of the half-life are listed in Table IV). The error is mainly related to the uncertainties of behavior of the β spectrum at low energies, the background model, the live time of measurements, the efficiency of selection of β events by the pulse-shape discrimination procedure to reject PMT noise, and the number of ^{113}Cd nuclei in the crystal.

TABLE IV. Different origins of uncertainties and their contributions to the half-life value of ^{113}Cd β decay.

Origin of the error	Range	Contribution to $T_{1/2}$ (10^{15} yr)
Statistical error	2403374 ± 1490 counts	±0.005
Number of ^{113}Cd nuclei	$8.858 \pm 0.014 \times 10^{22}$	±0.013
Live measuring time	2758 ± 6 h	±0.016
Efficiency of β events selection by pulse-shape discrimination procedure		$+0.017, -0.010$
Contribution of residual PMT noise	±1200 counts	±0.004
Detection efficiency of β decay (GEANT model uncertainty)	$99.97 \pm 0.02\%$	±0.002
Fit in different energy regions to reconstruct shape of the β spectrum under energy threshold		±0.027
Background model uncertainty		±0.024

Finally, we give the following half-life of ^{113}Cd relative to β decay:

$$T_{1/2} = (8.04 \pm 0.05) \times 10^{15}\text{yr}.$$

B. Shape of the ^{113}Cd β spectrum and Q_β value

Spin and parity in the β decay $^{113}\text{Cd} \rightarrow {}^{113}\text{In}$ (see Fig. 1) are changed from the initial $1/2^+$ to the final $9/2^+$ values ($\Delta J^{\Delta\pi} = 4^+$); such transition is classified as four-fold forbidden non-unique β decay. The energy distribution of the emitted electrons is given by the formula:

$$\rho(E) = wpF(E, Z)(Q_\beta - E)^2 \cdot C(w), \qquad (1)$$

where E is the kinetic energy of the electron, $w = E/m_e c^2 + 1$ is its full energy in the units of the electron mass, $p = \sqrt{w^2 - 1}$ is the electron momentum (in $m_e c$ units), $F(E, Z)$ is the Fermi function which accounts for the distortion of the spectrum due to the electric field of the nucleus and of the atomic shell, and Z is the atomic number of the daughter nucleus. The first term $wpF(Q_\beta - E)^2$ in Eq. (1) presents the shape of the allowed β decay, and $C(w)$ is a correction factor. For $\Delta J^{\Delta\pi} = 4^+$ transitions $C(w)$ is described by quite complex expressions [28] in polynomials in p and q (momenta of electron and

neutrino, respectively) up to 8-th degree with coefficients dependent on different nuclear matrix elements (NME). To avoid these complex expressions (with NMEs often unknown or dependent on theory), correction factors for the non-unique β decays are often approximated by the following formula:

$$C(w) = 1 + c_1/w + c_2 w + c_3 w^2$$

with c_i constants.

However, for the β decay of ^{113}Cd, starting from the work in Ref. [5], another expression is traditionally used for the correction factor:

$$C(w) = p^6 + 7a_1 p^4 q^2 + 7a_2 p^2 q^4 + a_3 q^6, \qquad (2)$$

where $q = (Q_\beta - E)/m_e c^2$ is the momentum of the emitted neutrino. Such an expression is characteristic of transitions with $\Delta J^{\Delta\pi} = 4^-$, i.e. for three-fold forbidden unique β decays; nevertheless it was successfully used in Refs. [5,6] for analysis of the ^{113}Cd spectrum.

We also use this dependence by fitting the measured experimental spectrum in the energy region 28–360 keV (see Sec. IV A) by convolution of the ideal β shape with the response function of the detector:

$$f(E) = \int_0^{Q_\beta} \rho(E')R(E, E')dE', \qquad (3)$$

where $\rho(E')$ is given by Eq. (1) with the correction factor $C(w)$ of Eq. (2), and $R(E, E')$ is a Gaussian,

$$R(E, E') = \frac{1}{\sqrt{2\pi}\sigma(E')} \exp\left(-\frac{(E - E')^2}{2\sigma^2(E')}\right),$$

with $\sigma(E) = \sqrt{6.8E}/2.3548$ established in calibration measurements and relevant in the energy region of interest (see Sec. II B).

The Fermi function was taken in the form

$$F(E, Z) = \text{const}\, p^{2s-2} \exp(\pi\eta)|\Gamma(s + i\eta)|^2,$$

where $s = \sqrt{1 - (\alpha Z)^2}$, $\eta = \alpha Z w/p$, $\alpha = 1/137.036$, and Γ is the gamma function. This is an approximation not accounting for finite nuclear size and screening by the atomic electrons; however, it accurately reproduces a more complex function which accounts for these effects [28] (with $<1\%$ deviation) for $Z = 49$ and in the given energy region.

The constants a_i and Q_β value were the free parameters of the fit (with Q_β entering as in the subintegral function $\rho(E)$ as well being the upper limit of integration). The result of the fit (χ^2/n.d.f. $= 191/162 = 1.18$) is shown in Fig. 7; the area under the theoretical curve in the whole energy region gave the value $S = 2\,399\,050$ events, fully consistent with the results of the previous section ($S = 2\,403\,374 \pm 1490$).

The obtained values of the a_i coefficients are the following:

$$\begin{aligned} a_1 &= (1.016 \pm 0.005), \\ a_2 &= (1.499 \pm 0.016), \\ a_3 &= (3.034 \pm 0.045). \end{aligned} \qquad (4)$$

The Q_β value was determined to be $Q_\beta = [344.9 \pm 0.2(\text{stat.}) \pm 21(\text{syst.})]$ keV, with the last value representing the energy resolution (σ) of the detector at energy 345 keV.

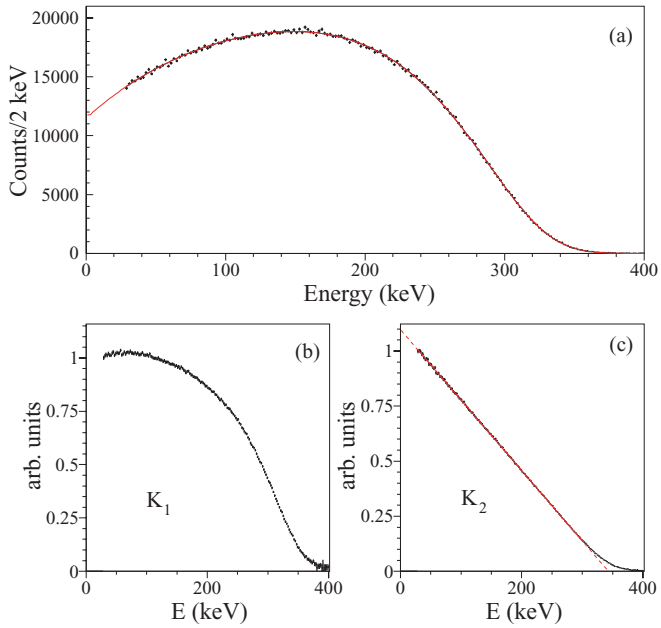

FIG. 7. (Color online) (a) Fit of the experimental spectrum (with background subtracted) by the convolution [Eq. (3)] of the ideal β shape [Eqs. (1) and (2)] with the detector response function. (b) Kurie plot for ^{113}Cd β decay, not accounting for correction factor $C(w)$. (c) Kurie plot, accounting for $C(w)$, and its fit by the linear function.

We have to note here that we were not able to reproduce the measured spectrum with the accepted value of $Q_\beta = (320 \pm 3)$ keV [2]. If the Q_β parameter in the fit was fixed at the accepted value, it resulted in the gap between the experimental spectrum and fitting curve at energies above $\simeq$250 keV which was not filled by changing other parameters of fit. We have observed such deviation from the accepted value systematically in all our measurements with $CdWO_4$ scintillators since 1989 (see, e.g, Refs. [6,12,16]).

Figure 7 shows the Kurie plots not accounting and accounting for the correction factor $C(w)$ calculated as

$$K_1(E) = \sqrt{\frac{N(E)}{pwF(E, Z)}}, \quad K_2(E) = \sqrt{\frac{N(E)}{pwF(E, Z)C(w)}},$$

respectively, where $N(E)$ is the number of events in the experimental spectrum at the energy E. As seen from the Fig. 7(c), the Kurie plot with $C(w)$ taken into account can be perfectly described by a straight line. Its fit in energy interval 28–300 keV (χ^2/n.d.f. $= 0.37$) gives the endpoint energy value of $Q_\beta = (343.1 \pm 0.6)$ keV.

C. Comparison with other experiments and theory

The half-life measured in the present experiment, $T_{1/2} = (8.04 \pm 0.05) \times 10^{15}$ yr, is in agreement with our previous value [6] as well as with results obtained in other experiments (see Table I).

Theoretical calculations for the ^{113}Cd half-life were absent until now. It was calculated only recently in the framework of the microscopic quasiparticle-phonon model (MQPM) as $T_{1/2} = 16.9 \times 10^{15}$ yr [29]. This value was further recalculated

in a proton-neutron variant of the MQPM as $T_{1/2} = 10.5 \times 10^{15}$ yr [30], in substantial agreement with the experimental values.

As for the shape of the ^{113}Cd spectrum, the following values for the a_i constants were determined in previous works [5,6]:

$$\begin{aligned} a_1 &= (0.765 \pm 0.095), \\ a_2 &= (0.589 \pm 0.177), \\ a_3 &= (2.04 \pm 0.74), \end{aligned} \tag{5}$$

$$\begin{aligned} a_1 &= (1.01 \pm 0.01), \\ a_2 &= (1.48 \pm 0.05), \\ a_3 &= (0.68 \pm 0.21). \end{aligned} \tag{6}$$

The experimental results [Eq. (4)] agree with our previous values [Eq. (6)] except for the a_3 parameter. This disagreement is understandable since a_3, being the coefficient at q^6, is important at high values of the neutrino momentum, i.e., at low electron energies. The energy threshold of 44 keV in the older experiment [6] was higher than the threshold achieved in the present measurements (28 keV), and the a_3 value was underestimated. Spectrum shapes obtained in different experiments are shown in Fig. 8(a), where we account also for the different experimental Q_β values: 319 [5], 337 [6], and 345 keV in the present study. The distributions are calculated in accordance with Eqs. (1) and (2) (i.e., without distortion due to finite energy resolution of a detector) and normalized to area equal to 1.

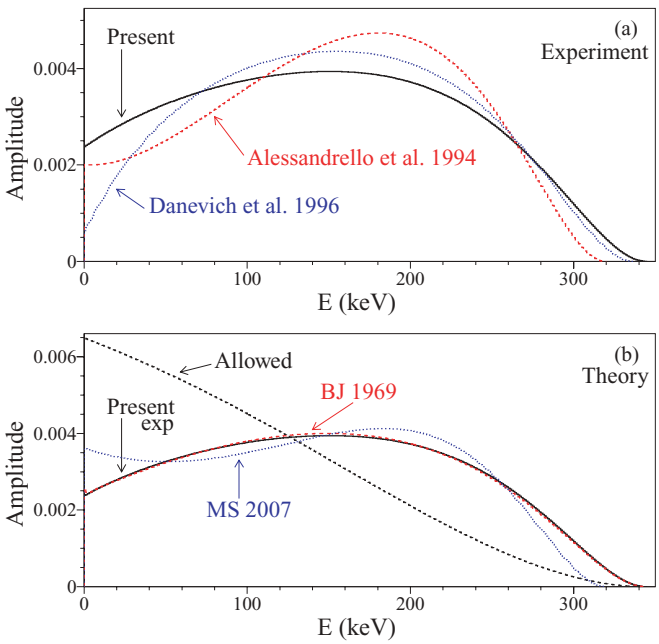

FIG. 8. (Color online) Energy spectra of electrons in the ^{113}Cd β decay: (a) experimental (without distortion due to finite energy resolution of the detector) and (b) theoretical. All spectra are normalized to area equal 1. Theoretical shape for the three-fold forbidden unique β decay ($\Delta J^{\Delta\pi} = 4^-$) calculated in accordance with Ref. [28] (curve BJ 1969) practically coincides with the experimental spectrum obtained in this work for the ^{113}Cd four-fold forbidden non-unique β decay ($\Delta J^{\Delta\pi} = 4^+$).

In fact, Eq. (2) for the correction factor of the three-fold forbidden unique β decay is a simplified version of the more complex expression [28]:

$$C(w) = q^6 + 7\lambda_2(w)q^4p^2 + 7\lambda_3(w)q^2p^4 + \lambda_4(w)p^6, \tag{7}$$

where $\lambda_i(w)$ are not constants but different Coulomb functions dependent on energy and calculated in Ref. [28]. The theoretical shape for ^{113}Cd with correction factor (7) with $\lambda_i(w)$ [28] is shown in Fig. 8(b) (curve BJ 1969). It practically coincides with the experimental shape obtained in this work. This is a very interesting fact because Eq. (7) represents the spectrum for the unique $\Delta J^{\Delta\pi} = 4^-$ transition, while in case of ^{113}Cd we have non-unique β decay with $\Delta J^{\Delta\pi} = 4^+$. Theoretical descriptions of these decays are very different: the energy spectrum in unique decays is determined by some energy-dependent function related to only one nuclear matrix element, while for non-unique decays the description is much more complex.

We also show in Fig. 8(b) the allowed shape calculated by means of Eq. (1) for $Z = 49$ [$C(w) = 1$]. As noted in Ref. [7] (where such a curve was referred to as being presented at the Web Table of Isotopes [31] for ^{113}Cd), it is very different from the experimentally observed spectrum. The curve MS 2007 is the theoretical spectrum recently calculated in the pnMQPM approach [30]; it takes into account that the β decay of ^{113}Cd is non-unique, and that the corresponding correction factor $C(w)$ has a much more complex expression than Eq. (7). In fact, it is the sum of polynomials in q and p up to the eighth degree with coefficients dependent on energy as well as on 12 different nuclear matrix elements; the latter ones were calculated in Ref. [29]. While it is not very far from our measurements, the character bend in this theoretical spectrum resembles the experimental shape obtained in Ref. [5]. In our opinion, further investigation of the ^{113}Cd spectrum shape (and Q_β value) would be desirable.

V. CONCLUSIONS

The low background $CdWO_4$ crystal scintillator with mass of 0.434 kg was used in the experiment underground at the LNGS over a 2758 h period to investigate the ^{113}Cd β decay. The deeper experimental site, the low background setup, the discrimination capability of the PMT noise and α events, the larger exposure, the lower level of background, and the slightly better energy resolution allowed us to improve previous results obtained in the older experiment with the same crystal [6]. The number of ^{113}Cd nuclei was determined with the precise mass spectrometric measurements of cadmium isotope abundances in the $CdWO_4$ crystal, i.e., with a higher accuracy than in the previous experiments. The background of the detector was reconstructed on the basis of a careful analysis both of the data of the low-background experiment, and of the ICP-MS measurements of the $CdWO_4$ contamination. All these improvements have allowed the determination of the spectral shape of the ^{113}Cd β decay and its half-life $T_{1/2} = (8.04 \pm 0.05) \times 10^{15}$ yr with a better accuracy than in other measurements.

Further progress in the investigation of ^{113}Cd β decay—in particular, more precise measurement of the spectrum shape—could be reached, e.g., with $CdWO_4$ crystal scintillators of higher quality (enriched and depleted in ^{113}Cd) and with a further decrease of the background.

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