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NUCLEI
Experiment

Beta Decay of ^{113}Cd

F. A. Danevich, A. Sh. Georgadze, V. V. Kobychev, B. N. Kropivyansky, V. N. Kuts, A. S. Nikolayko, O. A. Ponkratenko, V. I. Tretyak, and Yu. G. Zdesenko

Institute for Nuclear Research, National Academy of Sciences of Ukraine, pr. Nauki 47, Kiev, 252028 Ukraine

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Abstract – Beta decay of ^{113}Cd is investigated experimentally in the Solotvina underground laboratory. The shape of the spectrum of the nonunique fourth-forbidden β decay of ^{113}Cd is measured with a low-background scintillation setup based on a $CdWO_4$ crystal. The half-life is found to be $T_{1/2} = (7.7 \pm 0.3) \times 10^{15}$ yr.

1. INTRODUCTION

The ^{113}Cd nucleus is among the most long-lived radioactive nuclei existing in nature. Its decay is a nonunique fourth-forbidden β transition ($I^\pi = 1/2^+$ for ^{113}Cd and $9/2^+$ for ^{113}In with $\Delta I^{\Delta\pi} = 4^+$). The energy of the transition ^{113}Cd–^{113}In is 316 ± 4 keV [1].

Attempts at detecting the β activity of ^{113}Cd were made in 1950 and in 1962 [2, 3]. Because the sensitivity of the methods used in these studies proved insufficient, only limits on $T_{1/2}$ were established. Greth *et al.* [4], who used a proportional counter to measure the activities of two cadmium samples alternatively – one enriched in ^{113}Cd to 99.4% and the other with the natural content of ^{113}Cd (12.22%) – were the first to observe the β decay of ^{113}Cd. The detection efficiency was estimated using β particles from a ^{87}Rb source. The effect-to-background ratio in this experiment was very small (approximately 1/50). A value of $(9.3 \pm 1.9) \times 10^{15}$ yr was found in [4] for the ^{113}Cd half-life.

The objective of this study is to determine more accurately $T_{1/2}$ for ^{113}Cd and to measure the corresponding β-decay spectrum. This spectrum is of considerable interest for studying the form factor of the fourth-forbidden β transition because the β decay of ^{113}Cd occurs to the ground state and is not masked by transitions forbidden in lower orders.

First measurements of the β-spectrum shape for ^{113}Cd were reported in [5], where a low-background CdTe detector of volume 0.27 cm^3 was used. However, the reliability of this result is low because of a significant background, which defied all attempts at analysis in the corresponding energy range (this is evident from the uncertainty in determining $T_{1/2}$: $T_{1/2} = (4 - 12) \times 10^{15}$ yr). Moreover, the shape of the spectrum was distorted because of the absorption of β particles at the boundary between the sensitive and dead volumes of the detector. Recently, the β decay of ^{113}Cd was detected with a low-temperature calorimeter based on a $CdWO_4$ crystal (58 g) with a resolution of about 5 keV [6]. The half-life and the endpoint energy were found to be $T_{1/2} = (9.3 \pm 0.5 \pm 1) \times 10^{15}$ yr and $(318 \pm 1.4 \pm 5$ keV), respectively, and the shape of the β spectrum was determined. A high background of the detector in the low-energy section of the spectrum was the principal source of the systematic error in this experiment.

2. DETECTOR AND EXPERIMENTAL PROCEDURE

Scintillation crystals of $CdWO_4$ used successfully in searches for double β decay of ^{116}Cd [7] offer strong possibilities for investigating ^{113}Cd. The natural isotopic content of ^{113}Cd makes it possible to perform experiments with natural-cadmium crystals. These scintillators possess high energy resolution (it was found in [8] that δE is 9.5% for $CdWO_4$ of diameter 25 mm and height 27 mm irradiated by 662-keV photons from ^{137}Cs) and low content of radioactive impurities [7]. Owing to the latter, the intrinsic background of these crystals is low. The radiation maximum of $CdWO_4$ scintillators occurs near the point 490 nm; the deexcitation constant is 9 - 13 μm; the refractive index is equal to 2.2 - 2.3, and the density amounts to 7.9 - 8.1 g/cm^3. The application of $CdWO_4$ scintillators in studying the β decay of ^{113}Cd makes it possible to implement a procedure in which the same substance serves as the detector and the source. This ensures an almost 100% efficiency of electron detection.

To measure the β spectrum of ^{113}Cd, we used a $CdWO_4$ crystal of diameter 40 mm and height 45 mm (454.18 ± 0.05 g). To reduce the energy threshold and to improve resolution, the crystal was scanned by two FEU-110 photomultipliers (PEM) from both sides through polymethyl-methacrylate light guides of length 250 mm. The shielding against the external background consisted of fluoroplastic, oxygen-free copper, mercury, and lead of thickness 20, 25, 70, and 225 mm, respectively. The experiment was carried out in the Solotvina underground laboratory (Kiev Institute for Nuclear Research, National Academy of Sciences of Ukraine) arranged in a salt mine at a depth of 1000 m below sea level [9]. At this depth, the degree of suppression of the cosmic-muon flux is 10^5; the γ background

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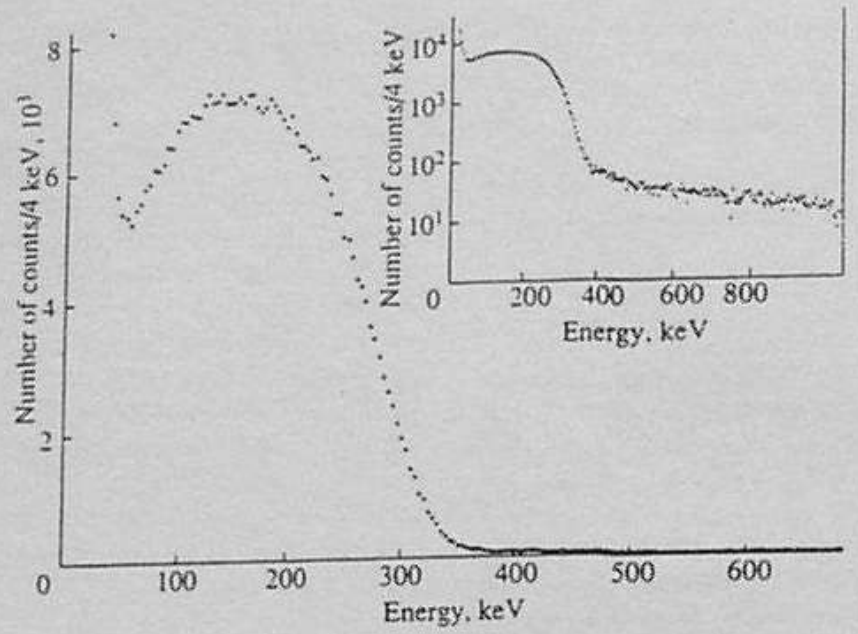

Fig. 1. Background spectrum of a $CdWO_4$ scintillation crystal of dimensions 40×45 mm^3 (time of measurement is 433.2 h).

is lower than in a usual above-ground laboratory by a factor of 30 - 70, and the thermal-neutron flux is below 2.7×10^{-6} cm^{-2} s^{-1}. The radon activity in air is constant (30 Bq/m^3). Signals from the dynodes of the PEM of the main detector were summed after formation and amplification and were transferred to the input of an analog-to-digital converter of a multichannel pulse analyzer. The detection of events was allowed only when both PEMs were simultaneously actuated. The coincidence system was adjusted in such a way as to reduce the number of PEM noise pulses and, at the same time, to ensure the maximum efficiency of detection of useful signals. Calibration measurements with ^{137}Cs (662-keV) and ^{241}Am (59.5-keV) sources were carried out in the modes of coincidences and simple summation. It was established that in the coincidence mode, no less than 98.4 and 99.4% of counts were detected at the ^{137}Cs and ^{241}Am peaks. The accumulated spectra were input into a computer for processing.

The background of $CdWO_4$ was measured for 433.2 h divided into three runs, each being of about a week in duration. In between, we performed the calibration of the spectrometer energy scale using ^{241}Am, ^{137}Cs, ^{22}Na, ^{207}Bi, ^{226}Ra, and ^{232}Th photon sources. The resolution (FWHM ΔE) with photons from these sources amounted to 25 keV (E_γ = 59.5 keV, ^{241}Am), 41 keV (239 keV, ^{232}Th), 49 keV (352 keV, ^{226}Ra), 62 keV (511 keV, ^{22}Na), 68 keV (609 keV, ^{226}Ra), 65 keV (570 keV, ^{207}Bi), and 71 keV (662 keV, ^{137}Cs). The dependence of the energy resolution on the energy E was approximated by the expression $\Delta E = \sqrt{a + bE}$ (keV), where $a = 131 \pm 0.94$ keV^2 and $b = 7.33 \pm 0.18$ keV. Owing to the stable functioning of the spectrometric channel during the entire cycle of measurements, the shift of calibration peaks was within the channel value (2 - 3 keV). Data analysis and processing were performed using SM-1420 and VAX 789/11 computers.

3. ANALYSIS OF RESULTS AND DETERMINATION OF THE ^{113}Cd HALF-LIFE

The spectrum of $CdWO_4$ background is shown in Fig. 1 on linear and logarithmic scales. In the energy range 55 - 380 keV, we clearly see a continuous distribution that corresponds to the β decay of ^{113}Cd and which is characteristic of a β spectrum. A sharp increase in the number of counts at energies below 55 keV is associated with PEM noises that were not suppressed by the coincidence system. A continuous exponential distribution in the energy range 380 - 1000 keV stems predominantly from external background and, possibly, from the decays of radioactive impurities in the crystal. A low level of background and the absence of distinct peaks in this section of the spectrum enabled us to estimate the probable contributions from the external and internal radioactivities to the area of the ^{113}Cd spectrum. The measured spectrum is characterized by a high (greater than 50/1) effect-to-background ratio.

To reduce the error in determining the half-life and to obtain a possibly more accurate shape of the β distribution, the contribution of PEM noises and the contribution of external detector background were subtracted from the experimental spectrum. We measured the spectrum of PEM noises in the setup for 22.4 h using a Plexiglas prototype of the appropriate size instead of the $CdWO_4$ crystal. The fraction of subtracted counts is

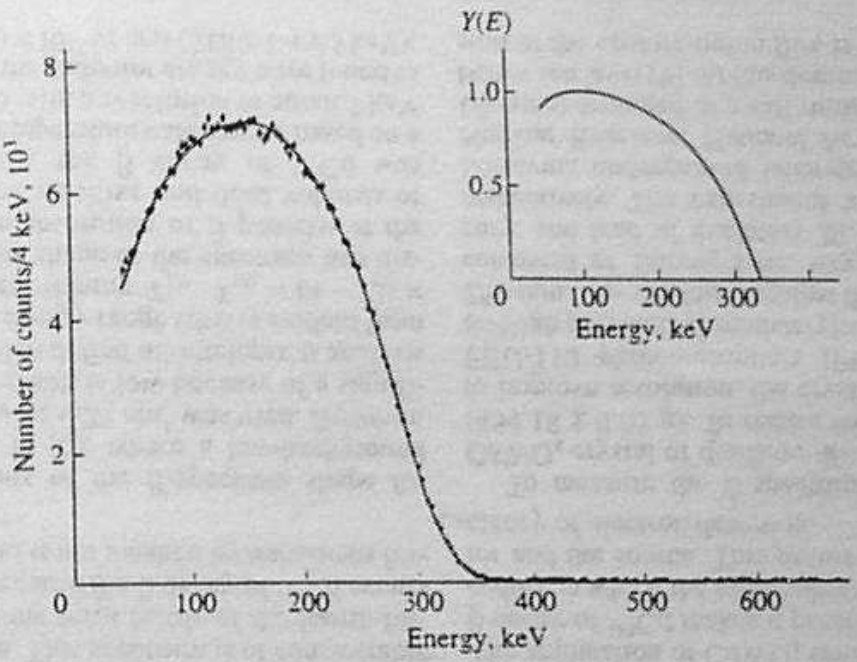

Fig. 2. ^{113}Cd β spectrum (points) due to the construction elements of the setup (these data were obtained by subtracting noises of photomultipliers and external background from the experimental spectrum shown in Fig. 1). The function $f(E)$ (curve) is the result of fitting the experimental spectrum. The Curie graph $Y(E) = [N(E)/F(Z, E)p(E + mc^2)]^{1/2}$ for the ^{113}Cd β spectrum is shown in the inset.

insignificant; in the range 44 - 380 keV, it amounts to 0.5% of the total number of counts.

With the aim of taking into account the possible intrinsic background of the $CdWO_4$ scintillator, we estimated the content of the most probable radioactive impurities of both natural and artificial origins. From analysis of the background spectrum in various energy ranges, we obtained the following constraints on activities (in mBq/kg): <3.7 for ^{40}K, <1.1 for ^{90}Sr, <7 for ^{106}Ru, <8 for ^{113m}Cd, <0.3 for ^{125}Sb, <0.2 for ^{134}Cs, <0.3 for ^{137}Cs, <0.1 for ^{226}Ra, <0.1 for ^{232}Th, and <0.3 for ^{238}U. If we take (very conservatively) these limits for the real content, the total contribution from all these impurities to the β spectrum of ^{113}Cd does not exceed 1.3%. However, the possibility that the crystal is contaminated with β emitters whose endpoint decay energies are not lower than 300 keV (for example, ^{14}C, ^{87}Rb, and ^{135}Cs) cannot be ruled out completely. Constraints on impurities of this kind were obtained by analyzing the background spectra of an enriched $^{116}CdWO_4$ crystal (of volume 11.4 cm^3 and with $\delta(^{116}Cd) = 83\%$) in which the content of ^{113}Cd is lower than in the main detector approximately by a factor of 6. For this purpose, we used the spectrum accumulated in 951 h in the same setup and under the same conditions as in the experiment with the natural $CdWO_4$ crystal. Assuming that the two detectors had approximately the same degree of purity (it was shown in [7] that this is the case for other impurities), we used the resulting constraints on the total counting rate due to pure β emitters with $E < 300$ keV (no more than 24 count/h in a 454-g detector at energies above 44 keV) to estimate the systematic error in determining ^{113}Cd activity.

To estimate external background, we used two approaches. In the first, the experimental spectrum in the range 380 - 1000 keV was approximated by an exponential curve, which was then extrapolated to the region of the β distribution. In the range 44 - 380 keV, the background contribution was 1.8%. In the second approach, the shape of the background spectrum was measured by the detector without passive shielding (the crystal was surrounded only by a thin fluoroplastic layer). The fraction of ^{113}Cd β decay in this spectrum was insignificant (about 3%). Following elimination of this fraction and normalization in the energy range 380 - 1000 keV, the resulting spectrum was used as a model of external background (in this case, the contribution of the external background to the β spectrum of ^{113}Cd amounts to 1.2%). The ^{113}Cd β spectrum due to construction elements of the setup was obtained by subtracting PEM noises and external background from the initial experimental spectrum (Fig. 1) and is shown in Fig. 2 by points. The sum of counts in the energy range 44 - 380 keV is $379560 \pm 620(\text{stat.})^{+2200}_{-12100}$ (syst.). As the spectral composition of the external γ background within the shielded volume can differ from that measured by the unshielded detector, we assumed that the actual background can be either twice as large or twice as small as the background in the model used. These values, as well as the possible contribution from radioactive impurities in the crystal, were taken into account in the systematic error.

The efficiency of detection of ^{113}Cd β decays was determined as the fraction of β particles whose energy losses in the detector exceed the detection threshold (44 keV). The spectrum of energy losses was calculated by the Monte Carlo method implemented in the GEANT3.15 code [10], the reconstructed shape of the β spectrum $N(E)$ [see formulas (1) and (2) below] being used for the energy distribution of β particles. To take into account possible deviations of the real spectrum from the approximation used, we considered two limiting cases of the behavior of the real β spectrum in the region 0 - 44 keV: the straight line that is tangential to the spectrum at the point 44 keV and the straight line that connects this point with the origin. The calculated detection efficiency is $(92.2^{+1.7}_{-2.9})$% (possible losses of signals in the coincidence mode of detection are included into the error in efficiency). Ultimately, the activity of ^{113}Cd in the crystal amounts to $0.2640^{+0.0084}_{-0.0097}$ Bq. When the number of ^{113}Cd nuclei that is equal to $(9.277 \pm 0.046) \times 10^{22}$ is taken into account, the value $T_{1/2} = (7.7 \pm 0.3) \times 10^{15}$ yr is obtained for the ^{113}Cd half-life with respect to β decay. This value does not contradict the results obtained in previous studies: $(9.3 \pm 1.9) \times 10^{15}$ yr [4] and $(9.3 \pm 0.5 \pm 1) \times 10^{15}$ yr [6]. However, our result is characterized by a smaller error due to a considerably greater effect-to-background ratio achieved in this experiment. The main contribution to the uncertainty in $T_{1/2}$ (about 70%) comes from the error that is introduced into the detection efficiency because of the absence of reliable information about the behavior of the β spectrum below 44 keV.

Distortions present in the ^{113}Cd β spectrum shown in Fig. 2 are associated with the energy resolution of detector (at 320 keV, it is about 50 keV). To obtain the true shape of the spectrum, it is necessary to perform the deconvolution of the experimental spectrum. The intrinsic spectral distribution cannot be reconstructed exactly, but an approximate solution to this problem is quite reliable in many particular cases. The approximate approach can be applied if the detector response function is known within the energy region of interest and if there are no factors (such as a high counting rate or the presence of intense γ peaks with the corresponding Compton distributions) that can distort or complicate this response function. As the above conditions were satisfied in this experiment, we used the approximate method to reconstruct the spectrum. The true shape of the β spectrum $N(E)$ was approximated by a model function. For the latter, we used the theoretical expression for the probability of the fourth-forbidden β transition:

$$N(E) = F(Z, E)p(E + mc^2)(E_0 - E)^2 S(E). \quad (1)$$

Here, $F(Z, E)$ is the Fermi function, p is the electron momentum, E is the electron kinetic energy, m is the electron mass, E_0 is the endpoint energy of the β spectrum, and $S(E)$ is the form factor. Under the assumption that nuclear charge is pointlike [11], the form factor can be approximated as

$$S(E) \sim p^6 + 7a_1p^4q^2 + 7a_2p^2q^4 + a_3q^6, \quad (2)$$

where the coefficients a_i are combinations of nuclear matrix elements, and q is the neutrino momentum. To reconstruct the intrinsic spectral distribution, we need the convolution of the model function $N(E)$ with the detector resolution function $R(E, E')$

$$f(E') = \int R(E, E')N(E)dE. \quad (3)$$

The detector resolution function measured using the calibration sources has the Gaussian form

$$R(E, E') = \frac{1}{\sqrt{2\pi}\sigma(E)} \exp\left(-\frac{(E - E')^2}{2\sigma^2(E)}\right), \quad (4)$$

where $\sigma(E) = \sqrt{(131 + 7.33E)}/2.35$ and E' is the energy determined by the detector. In this procedure, we assume that the function characterizing detector response to electrons emitted within the $CdWO_4$ crystal is identical to that for the detection of photons under the conditions of full absorption. Using the least-squares method, we fitted the function $f(E')$ to the experimental spectrum from which the background and PEM noises were subtracted. In doing this, we varied the coefficients a_i and the endpoint energy of the β spectrum. The fitting procedure was performed using the MINUIT code [12]. The function $f(E')$ obtained as the result of this fitting procedure is shown by the curve in Fig. 2 (the corresponding value of χ^2 is 0.88). The endpoint energy of the β spectrum was found to be $E_0 = 337.4 \pm 0.3$(stat.) ± 22(syst.) keV. The error in determining E_0 comes from spectrometer resolution, which amounts to 15% at this energy. The values of the coefficients a_i are as follows: $a_1 = 1.01 \pm 0.01$, $a_2 = 1.48 \pm 0.05$, and $a_3 = 0.68 \pm 0.21$. The Curie graph corresponding to the β decay of ^{113}Cd (see Fig. 2) clearly demonstrates a significant deviation from the straight line corresponding to the allowed β transition.

6. CONCLUSION

We determined the ^{113}Cd half-life with a higher precision than in the previous studies [4, 6]. Our result is $T_{1/2} = (7.7 \pm 0.3) \times 10^{15}$ yr. We also measured the shape of the β spectrum. Experimental errors in $T_{1/2}$ are due primarily to the uncertainty in the shape of the spectrum in the energy range 0 - 44 keV. The accuracy in determining the half-life was higher than in the previ-

ous studies because of a considerable increase in the effect-to-background ratio (50/1).

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