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Search for double- β decay processes in ¹⁰⁸Cd and ¹¹⁴Cd with the help of the low-background CdWO₄ crystal scintillator

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Abstract. The search for double- β processes in ¹⁰⁸Cd and ¹¹⁴Cd was realized by using data of the low-background experiment with the CdWO₄ crystal scintillator at the Gran Sasso National Laboratories of the INFN. New improved half-life limits on double-beta processes were established, in particular $T_{1/2}^{0\nu 2\varepsilon}(^{108}\text{Cd}) \ge 1.0(2.7) \times 10^{18} \text{ yr}, T_{1/2}^{2\nu 2\beta}(^{114}\text{Cd}) \ge 1.3(2.1) \times 10^{18} \text{ yr}, \text{ and } T_{1/2}^{0\nu 2\beta}(^{114}\text{Cd}) \ge 1.1(2.5) \times 10^{21} \text{ yr}$ at 90(68)% C.L.

PACS. 29.40.Mc Scintillation detectors – 23.40.-s β decay; double β decay; electron and muon capture

1 Introduction

The neutrinoless mode of double-beta decay $(0\nu 2\beta)$ is considered as a powerful tool to investigate neutrino properties. Even in the case of negative results double-beta decay experiments provide important information in the form of limits on neutrino Majorana mass, right-handed admixtures in weak interaction, existence of Majorons, and other effects beyond the standard model.

Experimental efforts are concentrated mainly on the processes with emission of two electrons. The two-neutrino $(2\nu) 2\beta^-$ decay was observed in 10 isotopes with half-lives in the range of 10^{18} – 10^{24} yr. Half-life limits at the level of 10^{23} - 10^{25} yr were set for the $0\nu 2\beta^-$ decay of several nuclei [1-3]. Results of searches for capture of two electrons from atomic shells (2ε) , electron capture with positron emission $(\varepsilon\beta^+)$, emission of two positrons $(2\beta^+)$ are comparatively modest. The most sensitive experiments provide half-life limits at the level of 10^{18} – 10^{21} yr [1]. It should be stressed that even the allowed two-neutrino mode of $(2\varepsilon, \varepsilon\beta^+, 2\beta^+)$ processes is still not observed¹. At the same time, search for neutrinoless $\varepsilon \beta^+$ and 2ε decays could be important to distinguish the mechanism of the $0\nu 2\beta$ decay (due to non-zero neutrino mass or to the right-handed admixtures in weak interactions) [5].

CdWO₄ crystals possess several unique properties required for high-sensitivity 2β decay experiments: low level of intrinsic radioactivity, good scintillation characteristics, and pulse-shape discrimination ability, which allow one to effectively reduce background. The Solotvina experiment to search for double-beta decay of cadmium and tungsten isotopes was carried out with the help of enriched in ¹¹⁶Cd cadmium tungstate crystal scintillators (¹¹⁶CdWO₄) [6]. Recently, the low-background CdWO₄ crystal scintillator with mass of 0.434 kg was used in the experiment at the Gran Sasso National Laboratories of the INFN to investigate the ¹¹³Cd beta decay [7]. CdWO₄ crystals contain several potentially 2β -decaying isotopes of Cd and W. In the present article we will consider 2β decay of ¹⁰⁸Cd (its natural abundance is $\delta = 0.89(3)\%$ [8] and energy release in 2β decay is $Q_{\beta\beta} = 272(6)$ keV [9]) and ¹¹⁴Cd ($\delta = 28.73(42)\%$, $Q_{\beta\beta} = 540(3)$ keV).

Experimental limits on 2ε decays of ¹⁰⁸Cd on the level of 10¹⁷ yr were set in the experiment with 454 g CdWO₄ scintillator with natural Cd composition in exposition during 433 h [6]. Currently the best limit on $2\nu 2\varepsilon$ decay ($\simeq 10^{18}$ yr) followed in 2003 from the first data of the COBRA experiment with the small semiconductor detectors Cd_{0.9}Zn_{0.1}Te ($\simeq 3$ g, 1117 h of data collection) and CdTe ($\simeq 6$ g, 1645 h) [10].

The best limits on two neutrino and neutrinoless channels of 2β decay of ¹¹⁴Cd were set in the experiment [6] by using the ¹¹⁶CdWO₄ crystal scintillator. The neutrinoless channel was restricted as $T_{1/2}^{0\nu2\beta}(^{114}Cd) \geq 2.5(4.1) \times 10^{20} \text{ yr}$

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¹ An indication for $2\beta^+$ decay processes in ¹³⁰Ba and ¹³²Ba was obtained in geochemical measurements [4]; however, this result has to be confirmed in a direct counting experiment.

at 90(68)% C.L., while for the two-neutrino decay the limit $T_{1/2}^{2\nu_2\beta}(^{114}\text{Cd}) \geq 6.0(9.3) \times 10^{17} \text{ yr}$ was set. It should be noted that the ¹¹⁴Cd abundance in the ¹¹⁶CdWO₄ crystal [6] was equal to 6.5%, which is $\simeq 4$ times lower than that in CdWO₄ produced from natural cadmium.

Results of searches for 2β decay processes in ¹⁰⁸Cd and ¹¹⁴Cd by using data of the measurements with the CdWO₄ crystal scintillator [7] are presented in this paper.

2 Experiment and data analysis

The CdWO₄ detector, experimental set-up, measurements and data analysis are described in detail in [7]. Here we outline the main features of the experiment.

A clear CdWO₄ crystal $40 \,\mathrm{mm}$ in diameter by $43 \,\mathrm{mm}$ in length (mass of 434 g) was used in the experiment. The CdWO₄ scintillator was fixed inside a cavity $\oslash 47 \times 59 \text{ mm}$ 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 coupled on opposite sides to two low radioactive EMI9265-B53/FL, 3" diameter photomultipliers (PMT). The detector was installed deep underground in the low-background DAMA/R&D set-up at the Gran Sasso National Laboratories of the INFN. The detector was surrounded by Cu bricks and sealed in a low radioactive air-tight Cu box continuously flushed with high-purity nitrogen gas to avoid presence of residual environmental radon. The Cu box was surrounded by a passive shield made of high-purity Cu, $10\,\mathrm{cm}$ of thickness, $15\,\mathrm{cm}$ of low radioactive lead, $1.5\,\mathrm{mm}$ of cadmium and 4 to 10 cm of polyethylene/paraffin to reduce the external background. The shield was contained inside a Plexiglas box, also continuously flushed by highpurity nitrogen gas.

An event-by-event data acquisition system records amplitude, arrival time, and pulse shape of events by a 1 GS/s 8 bit DC270 Transient Digitizer by Acqiris (adjusted to a sampling frequency of 20 MS/s) over a time window of 100 μ s.

The measurements were carried out in the energy range $\approx 0.03-0.7 \,\text{MeV}$ over 2758 h. The energy dependence of the detector energy resolution was measured with ²²Na, ¹³³Ba, ¹³⁷Cs, ²²⁸Th and ²⁴¹Am sources. The energy resolution can be described by the function: FWHM_{γ} = $\sqrt{6.8 \cdot E_{\gamma}}$, where E_{γ} is the energy of γ quanta, and FWHM_{γ} and E_{γ} are in keV.

The energy spectrum accumulated in the low-background DAMA/R&D set-up with the CdWO₄ detector is presented in fig. 1. The counting rate in the spectrum below the energy of $\approx 380 \text{ keV}$ is mainly provided by the β decay of ¹¹³Cd. Contributions to the background above the energy 380 keV from a possible internal contamination of the CdWO₄ scintillator (⁴⁰K, ⁶⁰Co, ⁹⁰Sr–⁹⁰Y, ^{113m}Cd, ¹³⁷Cs, β active nuclides from U/Th families), and from external γ -rays were simulated with the help of the GEANT4 package [11]. The initial kinematics of the particles emitted in the decay of nuclei was given by an event generator DECAY0 [12]. The fit of the background spectrum by



Fig. 1. (Color online) Energy spectrum of the CdWO₄ scintillator measured over 2758 h in the low-background set-up together with the model of the background (shown by solid line at $E > 380 \,\mathrm{keV}$) and its main components: β spectra of 40 K, 90 Sr $^{-90}$ Y, 113m Cd, and contribution from the external γ quanta from PMTs. An expected energy distribution of $2\nu 2\beta$ decay of 114 Cd with $T_{1/2}^{2\nu 2\beta} = 1.3 \times 10^{18} \,\mathrm{yr}$ (excluded at 90% of C.L.) is also shown.

this model in the energy interval 380–700 keV and main components of the background are shown in fig. 1. There are no clear peculiarities in the spectrum which could be ascribed to the internal trace contamination by radioactive nuclides. Therefore only limits were obtained on the activities of 40 K ($\leq 5\,\mathrm{mBq/kg}$), 60 Co ($\leq 0.4\,\mathrm{mBq/kg}$), $^{90}\mathrm{Sr}^{-90}\mathrm{Y}$ ($\leq 1\,\mathrm{mBq/kg}$), $^{113m}\mathrm{Cd}$ ($\leq 3\,\mathrm{mBq/kg}$), $^{137}\mathrm{Cs}$ ($\leq 0.3\,\mathrm{mBq/kg}$) [7]. The limits on activities of U/Th daughters evaluated with the help of time amplitude analysis and pulse shape discrimination between α particles and γ quanta (β particles) are even more strong (the equilibrium of the chains was assumed to be broken in the CdWO₄ crystal): $^{232}\mathrm{Th}$ ($\leq 0.03\,\mathrm{mBq/kg}$), $^{226}\mathrm{Ra}$ ($\leq 0.02\,\mathrm{mBq/kg}$), $^{210}\mathrm{Po}$ ($\leq 0.06\,\mathrm{mBq/kg}$).

3 Results and discussion

3.1 Search for double electron capture in ¹⁰⁸Cd

For the 2ν double electron capture in ¹⁰⁸Cd from the K shell, the total energy released in the detector is equal to $2E_K = 48.8 \text{ keV}$ (where $E_K = 24.4 \text{ keV}$ is the binding energy of the electrons on the K shell of palladium atoms). The detection of such a little energy deposit requires rather low-energy threshold. The sufficiently low-energy threshold of 28 keV was reached in the experiment [7] thanks to the high quality of the scintillator, a satisfactory light collection, and the rejection of PMT noise with the help of pulse shape discrimination.

The low-energy part of the background spectrum of the $CdWO_4$ detector, after the rejection of PMT noise



Fig. 2. (Color online) A part of the energy spectrum of the CdWO₄ scintillation crystal measured over 2758 h and its fit (see text). An expected peak from $2\nu 2K$ decay of ¹⁰⁸Cd corresponding to the half-life $T_{1/2}^{2\nu 2K} = 1.1 \times 10^{18}$ yr (excluded at 90% C.L.) is also shown by the dashed line.

and corrected for lost β (γ) events after the pulse shape discrimination (see [7] for details), is shown in fig. 2. There are no peculiarities in the spectrum which could be ascribed to the two-neutrino double electron capture from the K shell in ¹⁰⁸Cd. Therefore only lower halflife limits can be set according to formula: $\lim T_{1/2} =$ $N \cdot \eta \cdot t \cdot \ln 2 / \lim S$, where N is the number of potentially 2β unstable nuclei, η is the detection efficiency, t is the measuring time, and $\lim S$ is the number of events of the effect searched for which can be excluded at given confidence level (C.L.).

Two approaches were used to estimate the value of $\lim S$. In the first one (the so-called 1σ approach), we used the statistical uncertainty of the number of events registered in the energy interval 36-64 keV, which contains $\eta = 93\%$ of the expected peak. 218821 events were observed in the energy interval. Considering the number of ¹⁰⁸Cd nuclei in the crystal $N = 6.45 \times 10^{21}$, and the measurement time t = 2758 h, it gives the half-life limit $T_{1/2}^{2\nu 2K}(^{108}\text{Cd}) \geq 2.8 \times 10^{18} \text{ yr at } 68\% \text{ C.L.}$ In the second approach, the experimental energy distribution was fitted in the energy interval 30-100 keV by the sum of the function representing the β spectrum of ¹¹³Cd (see [7] for details) and the expected model function for $2\nu 2K$ decay (Gaussian at the energy of 48.8 keV and FWHM = 18 keV). The least-squares fit ($\chi^2/\text{n.d.f.} = 50/30 = 1.65$) gives the total area of the effect -30 ± 790 counts which corresponds (in accordance with the Feldman-Cousins procedure [13]) to $\lim S = 1269$ (760) counts at 90%(68%) C.L. Taking into account practically 100% registration efficiency for the whole curve in this case, one can calculate the half-life limit:

$$T_{1/2}^{2\nu 2K}(^{108}\text{Cd}) \ge 1.1(1.9) \times 10^{18} \text{ yr at } 90\%(68\%) \text{ C.L.}$$

The peak expected for the $2\nu 2K$ decay of ¹⁰⁸Cd, with the half-life of 1.1×10^{18} yr, is shown in fig. 2.

For the neutrinoless double electron capture in ¹⁰⁸Cd, all available energy (transferred to X-rays/Auger elec-

trons, gamma quanta or conversion electrons) will result in the peak at $Q_{\beta\beta} = 272 \text{ keV}$ value. The response function was simulated with the help of the GEANT4 package and the event generator DECAY0. We assume that all available energy was transferred to one γ quantum with the energy $Q_{\beta\beta} - 2E_K = 223 \text{ keV}$, which is the most pessimistic hypothesis from the point of view of the registration efficiency. In this case it has a minimal possible value ($\eta = 0.85$). The fit of the measured spectrum in the energy interval 100–320 keV by the sum of the function representing the β spectrum of ¹¹³Cd and of the expected $0\nu 2\varepsilon$ peak gives the following limit:

$$T_{1/2}^{0\nu 2\varepsilon}(^{108}\text{Cd}) \ge 1.0(2.7) \times 10^{18} \text{ yr at } 90\%(68\%) \text{ C.L.}$$

The sensitivity of the experiment to 2β processes in ¹⁰⁸Cd can be significantly improved by using CdWO₄ crystal scintillators enriched in ¹⁰⁸Cd and depleted in ¹¹³Cd.

3.2 Double- β decay of ¹¹⁴Cd

The energy spectrum accumulated in the low-background set-up over 2758 h was used to set a limit on the $0\nu 2\beta$ decay of ¹¹⁴Cd. A Gaussian peak at the energy 540 keV with FWHM = 61 keV is expected for the $0\nu 2\beta$ decay of ¹¹⁴Cd. There is no indication in the data for the peak searched for.

To estimate the value of lim S, the energy spectrum was fitted in the energy range 420–680 keV with the step of 10 keV (the bounds were varied as 420–460 and 650– 680 keV) by the model composed of the simulated background components (⁴⁰K, ⁶⁰Co, ⁹⁰Sr–⁹⁰Y, ^{113m}Cd, ¹³⁷Cs, β active U/Th nuclides, external γ 's from PMT) and the $0\nu 2\beta$ peak searched for. The best least-squares fit (χ^2 /n.d.f. = 11.1/9 = 1.23 achieved in the energy interval 450–670 keV) gives the total area of the effect -26 ± 39 counts which corresponds to lim S = 41(18) counts at 90%(68%) C.L. Taking into account 98% registration efficiency for two electrons emitted in the $0\nu 2\beta$ decay of ¹¹⁴Cd, and the number of ¹¹⁴Cd nuclei in the crystal (2.08×10^{23}), one can calculate the half-life limit:

$$T_{1/2}^{0\nu2\beta}(^{114}\text{Cd}) \ge 1.1(2.5) \times 10^{21} \text{ yr at } 90\%(68\%) \text{ C.L.}$$

The expected $0\nu 2\beta$ peak of ¹¹⁴Cd with the half-life of 2.5×10^{20} yr (the limit reported in the previous experiment [6]) is shown in fig. 3.

The following model was used to estimate the lower limit for the $2\nu 2\beta$ decay of ¹¹⁴Cd: the function to describe the beta spectrum of ¹¹³Cd (see [7]) plus the sum of the simulated background components (similar as in a case of the $0\nu 2\beta$ decay of ¹¹⁴Cd). The best fit $(\chi^2/n.d.f. = 77/88 = 0.87)$, achieved in the energy interval 386–582 keV, gives -83 ± 21551 events for the effect searched for. It yields 35268(21466) counts excluded at 90%(68%) C.L., and consequently, the following half-life limit for the $2\nu 2\beta$ decay of ¹¹⁴Cd:

$$T_{1/2}^{2\nu 2\beta}(^{114}\text{Cd}) \ge 1.3(2.1) \times 10^{18} \text{ yr at } 90\%(68\%) \text{ C.L.}$$



Fig. 3. Energy spectrum of the CdWO₄ scintillator measured over 2758 h. The solid line represents the fit of the data by the model of background (see text). The expected $0\nu 2\beta$ peak of ¹¹⁴Cd with half-life 2.5×10^{20} yr (corresponds to the previous limit reported in [6]) is shown.

Table 1. Half-life limits on 2β processes in ¹⁰⁸Cd and ¹¹⁴Cd.

Nuclide	Decay	Experimental $T_{1/2}$, yr at 90%(68%) C.L.	
	channel	Present work	Previous results
¹⁰⁸ Cd	$2\nu 2K$	$\geq 1.1(1.9) \times 10^{18}$	$\geq 1.0 \times 10^{18} \ [10]$
	$0\nu 2\varepsilon$	$\geq 1.0(2.7) \times 10^{18}$	$\geq 1.5(2.5) \times 10^{17} \ [6]$
114 Cd	$2\nu 2\beta$	$\geq 1.3(2.1) \times 10^{18}$	$\geq 6.0(9.3) \times 10^{17} [6]$
	$0\nu 2\beta$	$\geq 1.1(2.5) \times 10^{21}$	$\geq 2.5(4.1) \times 10^{20} \ [6]$

The energy distribution of the $2\nu 2\beta$ decay of ¹¹⁴Cd excluded at the 90% C.L. is shown in fig. 1.

All obtained limits are summarized in table 1 where the results of the most sensitive previous experiments are listed too. As one can see, the limit for $0\nu 2\varepsilon$ decay in 108 Cd, and those for the two-neutrino and neutrinoless 2β decay of 114 Cd are higher than the previous results.

It should be stressed that the sensitivity for $T_{1/2}^{0\nu_2\beta}$ on the level of 10^{21} yr was achieved previously only for fifteen 2β -decaying nuclei from the full list of 69 isotopes [1]. Despite ¹¹⁴Cd has a comparatively low-energy of $Q_{\beta\beta}$ (and therefore from this point of view is not much competitive in the sense of the sensitivity to the neutrino mass), this nucleus has a rather high isotopic abundance of $\delta = 29\%$. It allows to discuss large-scale experiments without very expensive enriched isotopes.

Moreover, disadvantage of low $Q_{\beta\beta}$ can be an advantage thanks to the suppression of the 2ν mode. The $2\nu 2\beta$ decay rate of ¹¹⁴Cd is strongly suppressed as compared with those of other nuclei due to low $Q_{\beta\beta}$. Therefore, the energy region of the $0\nu 2\beta$ signal will be free of the background produced by the $2\nu 2\beta$ events, which can reach this region due to the poor energy resolution of a detector [14]. The suppression of the 2ν mode would be especially important in the search for the $0\nu 2\beta$ decay with Majoron $(0\nu 2\beta M)$ emission, whose distribution is continuous, because in this case the $0\nu 2\beta M$ events will not be distinguished from the $2\nu 2\beta$ background even with the help of the high-energy resolution detector.

4 Conclusions

Data of the low-background experiment to study the β decay of ¹¹³Cd with the help of the low-background CdWO₄ crystal scintillator [7] were used to search for double-beta processes in ¹⁰⁸Cd and ¹¹⁴Cd.

A new improved limit was set for the neutrinoless double electron capture of ¹⁰⁸Cd as $T_{1/2}^{0\nu^{2\varepsilon}} \geq 1.0(2.7) \times 10^{18}$ yr at 90%(68%) C.L.

New improved limits were set for the double-beta decay of ¹¹⁴Cd. The $2\nu 2\beta$ and $0\nu 2\beta$ channels were restricted as $T_{1/2}^{2\nu 2\beta} \geq 1.3(2.1) \times 10^{18} \text{ yr}$ and $T_{1/2}^{0\nu 2\beta} \geq 1.1(2.5) \times 10^{21} \text{ yr}$, respectively.

These values are higher than those reached in the previous experiments [6,10] and demonstrate the possibility of a scintillation experimental technique to search for double-beta processes in 108 Cd and 114 Cd.

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