Search for 2β decay of ¹⁰⁶Cd with an enriched ¹⁰⁶Cd WO₄ crystal scintillator in coincidence with four HPGe detectors

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(Received 8 October 2015; revised manuscript received 27 January 2016; published 11 April 2016)

A radiopure cadmium tungstate crystal scintillator, enriched in ¹⁰⁶Cd to 66%, with mass of 216 g (¹⁰⁶Cd WO₄), was used to search for double- β decay processes in ¹⁰⁶Cd in coincidence with four ultra-low-background high-purity germanium detectors in a single cryostat. Improved limits on the double- β processes in ¹⁰⁶Cd have been set on the level of $10^{20} - 10^{21}$ yr after 13 085 h of data taking. In particular, the half-life limit on the two-neutrino electron capture with positron emission, $T_{1/2}^{2\nu\epsilon\beta^+} \ge 1.1 \times 10^{21}$ yr, has reached the region of theoretical predictions. With this half-life limit the effective nuclear matrix element for the $2\nu\epsilon\beta^+$ decay is bounded as $M_{\rm eff}^{2\nu\epsilon\beta^+} \le 1.1$. The resonant neutrinoless double-electron captures to the 2718-, 2741-, and 2748-keV excited states of ¹⁰⁶Pd are restricted at the level of $T_{1/2} \ge (8.5 \times 10^{20} - 1.4 \times 10^{21})$ yr.

DOI: 10.1103/PhysRevC.93.045502

I. INTRODUCTION

Experiments to search for neutrinoless double- β (0 $\nu 2\beta$) decay are considered as a promising way to study properties of neutrino and weak interactions and test the lepton number conservation [1–3]. In addition, the process can be mediated by right-handed currents in weak interaction, existence of massless (or very light) Nambu-Goldstone bosons (majorons), and many other effects beyond the standard model [1,3–5].

Experimental efforts are concentrated mainly on the double- β decay with emission of two electrons (see reviews [6-13]). The results of the experiments to search for double- β processes with decrease of nuclear charge—the capture of two electrons from atomic shells (2ε) , electron capture with positron emission ($\varepsilon\beta^+$), and emission of two positrons $(2\beta^+)$ —are substantially more modest (we refer reader to the reviews [6,7,14] and references 11-27 in Ref. [15]). Even the allowed two-neutrino mode of the double- β plus processes is not yet detected unambiguously: there are only indications on two-neutrino double-electron capture in 130 Ba [16,17] and 78 Kr [18] with the half-lives on the order 10^{20} to 10^{21} yr. At the same time a strong motivation to search for neutrinoless $\varepsilon\beta^+$ decay is related to the possibility of refining the mechanism of the $0\nu 2\beta^{-}$ decay when observed, whether it appears due to the Majorana mass of neutrino or due to the contribution of the right-handed admixtures in the weak interaction [19]. In addition, experimental data on the two-neutrino decay could be useful to improve theoretical calculations of the decay probability.

The isotope ¹⁰⁶Cd (energy of decay $Q_{2\beta} = 2775.39$ (10) keV [20], natural isotopic abundance $\delta = 1.25(6)\%$ [21]) is one of the most suitable nuclei to search for the double- β plus processes. In addition the isotope ¹⁰⁶Cd is favored to search for resonant $0\nu_{2\varepsilon}$ transitions to excited levels of ¹⁰⁶Pd when there is a coincidence between the released energy and the energy of an excited state [15,22]. The decay scheme of ¹⁰⁶Cd is presented in Fig. 1.

The nuclide ¹⁰⁶Cd is one of the most studied (see a detailed review of the previous investigations in Ref. [15]). At present there are three experiments in progress aiming at search for double- β decay of ¹⁰⁶Cd. The TGV-2 experiment in the Modane underground laboratory in France utilizes 32 planar HPGe detectors with 16 thin foils of enriched ¹⁰⁶Cd ($\delta = 75\%$) installed between the detectors. Recently new enriched foils with a higher isotopic concentration of ¹⁰⁶Cd $\delta = 99.6\%$ were installed. The sensitivity of the experiment is at the level of lim $T_{1/2} \sim 10^{20}$ yr [24–26]. The main advantage of the TGV-2 experiment is a high sensitivity to the $2\nu 2K$ decay of ¹⁰⁶Cd with the theoretically shortest half-life of the three allowed channels.

The COBRA experiment uses an array of CdZnTe roomtemperature semiconductors $\approx 1 \text{ cm}^3$ each at the Gran Sasso underground laboratory in Italy. The sensitivity of the experiment to the double- β processes in ¹⁰⁶Cd is at the level of lim $T_{1/2} \sim 10^{18}$ yr [27].

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FIG. 1. Simplified decay scheme of ¹⁰⁶Cd [23] (levels at 2283–2714 keV are omitted). Energies of the excited levels and of the emitted γ quanta are in keV. Relative intensities of γ quanta are given in parentheses.

The third experiment, also carried out at the Gran Sasso laboratory, utilizes a radiopure cadmium tungstate crystal scintillator with mass 216 g produced from cadmium enriched in ¹⁰⁶Cd to 66% (¹⁰⁶Cd WO₄) [28]. At the first stage of the experiment the sensitivity at the level of $\lim T_{1/2} \sim$ 10^{20} to 10^{21} yr was reached for different channels of double- β decay of ¹⁰⁶Cd [15]. To increase the experimental sensitivity to the 2 β processes with emission of γ quanta, the ¹⁰⁶Cd WO₄ scintillator was placed inside a low-background HPGe detector with four Ge crystals. Here we report the final results of the experiment with the ¹⁰⁶Cd WO₄ scintillation detector operated in coincidence (anticoincidence) with the four-crystal HPGe γ detector. Preliminary results of the experiment were presented in conference proceedings [29–32].

II. THE EXPERIMENT

The ¹⁰⁶Cd WO₄ crystal scintillator was viewed through a lead tungstate (PbWO₄) crystal light guide ($\oslash 40 \times 83$ mm) by 3-in. low-radioactive photomultiplier tube (PMT) Hamamatsu R6233MOD (see Fig. 2). The PbWO₄ crystal was developed from deeply purified [33] archaeological lead [34]. The detector was installed in an ultra-low-background GeMulti HPGe γ spectrometer of the STELLA (SubTErranean Low Level Assay) facility [35] at the Gran Sasso underground laboratory of the INFN (Italy) at the depth of 3600 m of water equivalent. Four HPGe detectors of the GeMulti setup are mounted in one cryostat with a well in the center. The volumes of the HPGe detectors are approximately 225 cm³ each.

An event-by-event data acquisition system is based on two four-channel digital spectrometers (DGF Pixie-4, XIA,



FIG. 2. Low background ¹⁰⁶Cd WO₄ crystal scintillator (1) viewed through PbWO₄ light-guide (2) by PMT (3). The scintillator is installed between HPGe detectors (4). Scheme of the electronic chain: (PA) preamplifiers; (FAN IN/OUT) linear FAN-IN/FAN-OUT; (SST-09) home-made electronic unit to provide triggers for cadmium tungstate scintillation signals; (Pixie-4) four-channel all digital spectrometers; (PC) personal computer.

LLC). One device [marked (1) in Fig. 2] is used to provide spectrometric data for the HPGe detectors, while the second Pixie-4 (2) acts as a 14-bit waveform digitizer to acquire signals from the ¹⁰⁶Cd WO₄ detector at the rate of 18.8 MSPS over a time window 54.8 μ s. The second Pixie-4 unit records also trigger signals from the homemade unit SST-09, which provides the triggers only if the signal amplitude in the ¹⁰⁶Cd WO₄ detector exceeds ~0.6 MeV to avoid acquisition of a large amount of data caused by the decays of ¹¹³Cd^m ($Q_{\beta} = 586$ keV) present in the ¹⁰⁶Cd WO₄ crystal [15,36]. The signals from the timing outputs of the HPGe detectors after summing are fed to the third input of the second Pixie-4 digitizer to select coincidence between the ¹⁰⁶Cd WO₄ and HPGe detectors off line.

The ¹⁰⁶Cd WO₄ and HPGe detectors were calibrated with ²²Na, ⁶⁰Co, ¹³⁷Cs, and ²²⁸Th γ sources. The energy resolution of the ¹⁰⁶Cd WO₄ detector can be described by the function FWHM = $(21.7 \times E_{\gamma})^{1/2}$, where FWHM and E_{γ} are given in keV. The energy resolution of the HPGe detectors during the experiment was FWHM $\approx 2-3$ keV for the 1332-keV γ quanta of ⁶⁰Co.

Energy spectrum and distribution of the start positions of the ¹⁰⁶Cd WO₄ detector pulses relatively to the HPGe signals accumulated with the ²²Na γ source (see upper panel in Fig. 3) demonstrate presence of coincidences between the ¹⁰⁶Cd WO₄ and HPGe detectors under the condition that the energy of events in the HPGe detectors is equal to 511 keV (energy of annihilation γ quanta), while practically there is



FIG. 3. Energy spectra of ²²Na (upper figure), ¹³⁷Cs (middle figure), and ²²⁸Th (lower figure) γ sources accumulated by the ¹⁰⁶Cd WO₄ detector: with no coincidence (black circles) and in coincidence with energy 511 keV in the HPGe detector (red crosses). The data simulated by using the EGS4 Monte Carlo code are drawn by solid lines. (Inset) Distribution of the ¹⁰⁶Cd WO₄ detector pulses start positions relatively to the HPGe signals with the energy 511 keV accumulated with ²²Na source (the time shift of the ¹⁰⁶Cd WO₄ signals by \simeq 220 channels is due to the tuning of the digitizer to provide baseline data).

no coincidence in the data accumulated with ¹³⁷Cs. As one can see in Fig. 3, the distributions simulated by the EGS4 code [37] are in agreement with the experimental data obtained with ²²Na, ¹³⁷Cs, and ²²⁸Th γ sources. It should be stressed that the energy threshold of the ¹⁰⁶Cd WO₄ detector in the coincidence mode (\approx 50 keV) is much lower than that in the anticoincidence mode since the data acquisition in the coincidence mode is triggered by the signals in the HPGe detectors.

III. RESULTS AND DISCUSSION

A. Data analysis

The mean-time pulse-shape discrimination method (see, e.g., Ref. [38]) was used to discriminate $\gamma(\beta)$ events from α events caused by internal contamination of the crystal by uranium and thorium. The scatter plot of the mean time versus energy of the background events accumulated over 571 h by the ¹⁰⁶Cd WO₄ detector is depicted in Fig. 4. The efficiency of the pulse-shape discrimination is worse than that in the previous experiment [15] due to the lower light collection efficiency with the PbWO₄ light guide. Nevertheless, the distribution of the mean time for the events with energies in the range 0.9–1.1 MeV (see inset in Fig. 4) justifies pulse-shape discrimination between α particles and γ quanta (β particles). The mean time for γ quanta was measured with the ²²⁸Th γ



FIG. 4. Mean time (see text) vs the energy accumulated over 571 h with the ¹⁰⁶Cd WO₄ crystal scintillator in the low-background setup. The plus-minus two- σ interval for mean-time values corresponding to γ quanta (β particles) and one- σ interval for α particles are depicted. Events with the mean time values greater than $\approx 1.1 \times 10^4$ can be explained by the overlap of events (mainly of β decays of ¹¹³Cd^m in the crystal). Inset: Distribution of the mean times in the energy interval 0.9–1.1 MeV demonstrates the ability of pulse-shape discrimination between $\gamma(\beta)$ and α events.

source in the energy range 0.5–2.6 MeV as $\tau_{\gamma} = 9843$. The energy dependence of the mean-time distribution σ ($\sigma_{\gamma}^{\rm t}$) was determined as $\sigma_{\gamma}^{\rm t} = 3376/\sqrt{0.1 \times E_{\gamma}}$, where E_{γ} is energy of γ quanta in keV. The pulse-shape discrimination was also used to reject overlapped pulses (mainly caused by pileups of the ¹¹³Cd^m β decay events) and pileups of the PbWO₄ scintillation signals (characterized by rather short decay time at the level of $\approx 0.3 \,\mu s$ [38]) with the ¹⁰⁶Cd WO₄ signals (effective average decay time $\approx 13 \,\mu s$ [39]).

The energy spectra accumulated over 13 085 h by the ¹⁰⁶Cd WO₄ detector in anticoincidence with the HPGe detectors, in coincidence with event(s) in at least one of the HPGe detectors with energy E > 200 keV, E = 511 keV, and E = 1160 keV, are presented in Fig. 5. The events in the anticoincidence spectrum were selected by using the following cuts: (1) there is no signal(s) in the HPGe detectors, (2) the mean time (τ) value of a scintillation pulse is within the plus-minus two- σ interval around the central value: $\tau_{\gamma} - 2\sigma_{\gamma}^{\tau} < \tau < \tau_{\gamma} + 2\sigma_{\gamma}^{\tau}$ (the interval is shown in Fig. 4). The pulse-shape discrimination cut selects 95.5% of $\gamma(\beta)$ events. The spectrum of the ¹⁰⁶Cd WO₄ detector in coincidence with the energy release in at least one of the HPGe detectors of more than 200 keV was built with the help of the same pulse-shape discrimination cut. The energy spectra of the 106 Cd WO₄ detector in coincidence with the signal(s) in the HPGe detectors with energy 511 keV (1160 keV) were built by using the following cuts: (1) there is event(s) in at least one of the HPGe detectors with energy $E = 511 \pm 3\sigma_{511}$ keV $(E = 1160 \pm 2.3\sigma_{1160} \text{ keV})$ where σ_{511} and σ_{1160} are the energy resolutions of the HPGe detectors for the annihilation



FIG. 5. Energy spectra of γ and β events accumulated over 13 085 h by the ¹⁰⁶Cd WO₄ detector in anticoincidence with the HPGe detectors ("AC"), in coincidence with event(s) in at least one of the HPGe detectors with the energy E > 200 keV ("CC > 200 keV"), $E = 511 \pm 3\sigma_{511}$ keV ("CC 511 keV"), and $E = 1160 \pm 2.3\sigma_{1160}$ keV ("CC 1160 keV").

peak and for γ quanta with energy 1160 keV, respectively; (2) the signals in the ¹⁰⁶Cd WO₄ and HPGe detectors coincide within the plus-minus three- σ time interval (see inset of Fig. 3, the time cut selects 99.7% of the coinciding events); (3) the mean time value of a ¹⁰⁶Cd WO₄ scintillation pulse is within the $\pm 3\sigma_{\gamma}^{\rm T}$ interval. The pulse-shape discrimination cut selects 99.7% of the $\gamma(\beta)$ events.

The data accumulated by the ¹⁰⁶Cd WO₄ detector in anticoincidence with the HPGe detectors confirmed the assumption about surface contamination of the ¹⁰⁶Cd WO₄ crystal by ²⁰⁷Bi [15]. The γ peaks of ²⁰⁷Bi observed in Ref. [15] disappeared after the cleaning of the scintillator by potassiumfree detergent and ultrapure nitric acid.

The anticoincidence spectrum was fitted in the energy interval 940–3980 keV (χ^2 /n.d.f. = 157/118 = 1.33, where n.d.f. is the number of degrees of freedom) by the model built from the energy distributions simulated by EGS4 code [37]. The model includes radioactive contamination of the ¹⁰⁶Cd WO₄ crystal scintillator [15,36], external γ quanta from the materials of the setup (⁴⁰K, ²³²Th, ²³⁸U in the cryostat of the HPGe detector, PMT, PbWO₄ light guide, ²⁶Al in the aluminium well of the cryostat), distribution of α particles (which passed the pulse-shape discrimination cut to select β and γ events), cosmogenic ¹¹⁰Ag^m in the ¹⁰⁶Cd WO₄ scintillator, and two-neutrino $2\beta^-$ decay of ¹¹⁶Cd with the half-life $T_{1/2} = 2.62 \times 10^{19}$ yr [40] present in the ¹⁰⁶Cd WO₄ crystal with the isotopic abundance $\delta = 1.5\%$ [28]. The result of the fit and the main components of the background are shown in Fig. 6.

The energy spectrum accumulated by the HPGe detector is presented in Fig. 7 together with the background data taken over 4 102 h. The counting rate of the HPGe detector with the ¹⁰⁶Cd WO₄ detector inside exceeds slightly the background counting rate. Some excess (at the level of 30–170% depending on the energy of γ quanta) is observed in the peaks of ²¹⁴Bi



FIG. 6. The energy spectrum of the γ and β events accumulated over 13 085 h in the low background setup with the ¹⁰⁶Cd WO₄ crystal scintillator (points) together with the background model (red continuous superimposed line). The main components of the background are shown: the distributions of internal and external ("ext γ ") ⁴⁰K, ²³²Th, and ²³⁸U, distribution of residual α particles (α), cosmogenic ¹¹⁰Ag^m in the ¹⁰⁶Cd WO₄ scintillator (with activity 0.3 mBq/kg), and $2\nu 2\beta^{-}$ decay of ¹¹⁶Cd. The excluded distribution of the $0\nu\varepsilon\beta^{+}$ decay of ¹⁰⁶Cd to the ground state of ¹⁰⁶Pd with the half-life $T_{1/2} = 1.5 \times 10^{21}$ yr is shown too.



FIG. 7. Energy spectrum accumulated over 13 085 h by the low background HPGe γ detector with the ¹⁰⁶Cd WO₄ scintillation detector inside (solid red histogram) and background data measured over 4 102 h (blue dots). The energy spectra in the 200–400 keV (middle part) and 450–650 keV (lower part) energy intervals. Energies of γ quanta are in keV.



FIG. 8. Simulated response functions of the ¹⁰⁶Cd WO₄ detector to 2ε , $\varepsilon\beta^+$, and $2\beta^+$ processes in ¹⁰⁶Cd without coincidence (solid histograms) and in coincidence with 511 annihilation γ quanta in the HPGe detector (dotted histograms). Also the response functions of the ¹⁰⁶Cd WO₄ detector to the $2\nu2\varepsilon$ and $2\nu\varepsilon\beta^+$ decays to the 1134-keV excited level of ¹⁰⁶Pd in coincidence with 622-keV γ quanta in the HPGe detector are shown by dashed histograms.

and ²¹⁴Pb (daughters of ²²⁶Ra from the ²³⁸U family). We assume that the excess is due to the radioactive contamination of the ¹⁰⁶Cd WO₄ detector and due to the HPGe detector passive shield modification (some part of the passive shield was removed to install the ¹⁰⁶Cd WO₄ detector). We have also observed γ quanta with energy 1809 keV, which we ascribe to cosmogenic ²⁶Al in the aluminium well of the cryostat. Besides, there is a clear peak at the energy 262.5(5) keV with the counting rate 0.381(4) counts/h (the peak is shown in the middle part of Fig. 7). We ascribe the peak to γ quanta of the isomeric transition of ¹¹³Cd^m with energy 263.7(3) keV [41].

B. Limits on 2β processes in ¹⁰⁶Cd

The response functions of the ¹⁰⁶Cd WO₄ and HPGe detectors to the 2β processes in ¹⁰⁶Cd were simulated with the help of the EGS4 code. The simulated energy distributions in the ¹⁰⁶Cd WO₄ detector without coincidence and in coincidence with 511-keV γ quanta (in coincidence with 622-keV γ quanta for the $2\nu 2\varepsilon$ and $2\nu \varepsilon \beta^+$ decays to the 1134-keV excited level of ¹⁰⁶Pd) in the HPGe detector are presented in Fig. 8.

There are no peculiarities in the data accumulated with the ¹⁰⁶Cd WO₄ and HPGe detectors that could be ascribed to the 2β processes in ¹⁰⁶Cd. Therefore only lower half-life limits can be set by using the formula:

$$\lim T_{1/2} = (\ln 2) N \eta t / \lim S,$$
 (1)

where N is the number of ¹⁰⁶Cd nuclei in the ¹⁰⁶Cd WO₄ crystal ($N = 2.42 \times 10^{23}$), η is the detection efficiency, t is

the time of measurements, and $\lim S$ is the number of events of the effect searched for, which can be excluded at a given confidence level (CL). All the limits are presented in this paper with 90% CL.

We have analyzed different data to estimate limits on the 2β processes in ¹⁰⁶Cd. For instance, to derive the value of $\lim S$ for the $0\nu\varepsilon\beta^+$ decay of ¹⁰⁶Cd to the ground state of ¹⁰⁶Pd, the ¹⁰⁶CdWO₄ anticoincidence spectrum was fitted by the model built from the components of the background and the effect searched for. The best fit, achieved in the energy interval 1000-3200 keV, gives the area of the effect $S = 27 \pm 49$ counts, thus providing no evidence for the effect. In accordance with the Feldman-Cousins procedure [42], this corresponds to $\lim S = 107$ counts. Taking into account the detection efficiency within the interval given by the Monte Carlo simulation (69.3%) and the 95.5% efficiency of the pulse-shape discrimination to select γ and β events, we got the half-life limit: $T_{1/2} \ge 1.5 \times 10^{21}$ yr. The excluded distribution of the $0\nu\varepsilon\beta^+$ decay of ¹⁰⁶Cd to the ground state of ¹⁰⁶Pd is shown in Fig. 6. The limit is lower than that obtained in the previous stage of the experiment [15] due to slightly higher background in the high-energy part and worse energy resolution of the 106 Cd WO₄ detector. The counting rate of the 106 Cd WO₄ detector is substantially

The counting rate of the ¹⁰⁶Cd WO₄ detector is substantially suppressed in coincidence with the energy 511 keV in the HPGe detectors. The coincidence energy spectrum of the ¹⁰⁶Cd WO₄ detector is presented in Fig. 9. There are only 115 events in the energy interval 0.05–4 MeV, while the simulated background model (built by using the parameters of the anticoincidence spectrum fit) contains 108 counts. We have estimated values of lim *S* for the 2β processes in ¹⁰⁶Cd in different energy intervals. For instance, there are 62 counts in the energy interval 250–1600 keV, while the background model contains 68 counts. According to Ref. [42], one should take lim *S* = 8.8 counts. Taking into account the detection

FIG. 9. Energy spectrum of the ¹⁰⁶Cd WO₄ detector in coincidence with 511-keV annihilation γ quanta in at least one of the HPGe detectors (filled circles) acquired over 13 085 h. The excluded distributions of different 2β processes in ¹⁰⁶Cd are shown by different lines.

TABLE I. Half-life limits on 2β processes in ¹⁰⁶Cd. The experimental selection (AC, anticoincidence; CC, in coincidence, at the given energy with HPGe, HPGe using only the HPGe detectors) is also reported. The results of the most sensitive previous experiments are given for comparison.

Decay channel	Decay mode	Level of ¹⁰⁶ Pd (keV)	$T_{1/2}$ limit (yr) at 90% CL	
			Present work (Data)	Best previous limit
2ε	2ν	g.s.		$\geq 4.2 \times 10^{20}$ [25]
2ε	2ν	2+ 512	$\geq 9.9 \times 10^{20} \text{ (CC 512 keV)}$	$\geq 1.2 \times 10^{20}$ [25]
2ε	2ν	2+ 1128	$\geq 5.5 \times 10^{20} \text{ (CC 616 keV)}$	$\geq 4.1 \times 10^{20}$ [15]
2ε	2ν	0+ 1134	≥1.0 × 10 ²¹ (CC 622 keV)	$\geq 1.7 \times 10^{20}$ [15]
2ε	2ν	2+ 1562	$\geq 7.4 \times 10^{20} \text{ (CC 1050 keV)}$	$\geq 5.1 \times 10^{19}$ [15]
2ε	2ν	0+ 1706	$\geq 7.1 \times 10^{20} (\text{CC 1194 keV})$	$\geq 1.1 \times 10^{20}$ [15]
2ε	2ν	0+ 2001	$\geq 9.7 \times 10^{20} (\text{CC 873 keV})$	$\geq 2.9 \times 10^{20}$ [15]
2ε	2ν	0+ 2278	≥1.0 × 10 ²¹ (CC 1766 keV)	$\geq 1.6 \times 10^{20}$ [15]
2 <i>K</i>	0ν	g.s.	$\geq 4.2 \times 10^{20}$ (HPGe)	$\geq 1.0 \times 10^{21}$ [15]
LK	0ν	g.s.	$\geq 1.3 \times 10^{21}$ (HPGe)	$\geq 1.0 \times 10^{21}$ [15]
2L	0ν	g.s.	\geq 5.4 × 10 ²⁰ (HPGe)	$\geq 1.0 \times 10^{21}$ [15]
2ε	0ν	2^+ 512	$\geq 3.9 \times 10^{20} (\text{CC 512 keV})$	$\geq 5.1 \times 10^{20}$ [15]
2ε	0ν	2+ 1128	$\geq 5.1 \times 10^{20} \text{ (CC 616 keV)}$	$\geq 3.1 \times 10^{20}$ [15]
2ε	0ν	0+ 1134	$\geq 1.1 \times 10^{21} (CC \ 622 \ keV)$	$\geq 3.5 \times 10^{20}$ [15]
2ε	0ν	2+ 1562	$\geq 7.3 \times 10^{20} (\text{CC 1050 keV})$	$\geq 3.5 \times 10^{20}$ [15]
2ε	0ν	0+ 1706	$\geq 1.0 \times 10^{21} (\text{CC 1194 keV})$	$\geq 2.5 \times 10^{20}$ [15]
2ε	0ν	0+ 2001	$\geq 1.2 \times 10^{21} (CC \ 873 \ keV)$	$\geq 2.3 \times 10^{20}$ [15]
2ε	0ν	$0^+ 2278$	$\geq 8.6 \times 10^{20} \text{ (CC 1766 keV)}$	$\geq 2.1 \times 10^{20} [15]$
Res. 2 <i>K</i>	0ν	2718	$\geq 1.1 \times 10^{21} (\text{CC } 1160 \text{ keV})$	$\geq 4.3 \times 10^{20}$ [15]
Res. KL_1	0ν	4+ 2741	$\geq 8.5 \times 10^{20}$ (HPGe)	$\geq 9.5 \times 10^{20}$ [15]
Res. KL_3	0ν	2,3 ⁻ 2748	$\geq 1.4 \times 10^{21} \text{ (CC 2236 keV)}$	≥4.3 × 10 ²⁰ [15]
$arepsiloneta^+$	2ν	g.s.	$\geq 1.1 \times 10^{21} (\text{CC 511 keV})$	$\geq 4.1 \times 10^{20}$ [45]
$\varepsilon \beta^+$	2ν	2+ 512	$\geq 1.3 \times 10^{21} (\text{CC 511 keV})$	$\geq 2.6 \times 10^{20}$ [45]
$\varepsilon \beta^+$	2ν	$2^+ 1128$	$\geq 1.0 \times 10^{21} (\text{CC 511 keV})$	$\geq 3.1 \times 10^{20} \ [15]$
$arepsiloneta^+$	2v	0+ 1134	$\geq 1.1 \times 10^{21} (\text{CC 622 keV})$	$\geq 3.7 \times 10^{20}$ [15]
$arepsiloneta^+$	0ν	g.s.	$\geq 1.5 \times 10^{21} (AC)$	$\geq 2.2 \times 10^{21} \ [15]$
$\varepsilon \beta^+$	0ν	2+ 512	$\geq 1.9 \times 10^{21} (\text{CC 511 keV})$	$\geq 1.3 \times 10^{21} \ [15]$
$arepsiloneta^+$	0ν	$2^+ 1128$	$\geq 1.3 \times 10^{21} \text{ (CC 511 keV)}$	$\geq 5.7 \times 10^{20}$ [15]
ϵeta^+	0ν	0+ 1134	$\geq 1.9 \times 10^{21} (\text{CC 622 keV})$	$\geq 5.0 \times 10^{20}$ [15]
$2\beta^+$	2ν	g.s.	$\geq 2.3 \times 10^{21} (\text{CC 511 keV})$	$\geq 4.3 \times 10^{20}$ [15]
$2\beta^+$	2ν	2+ 512	$\geq 2.5 \times 10^{21} \text{ (CC 511 keV)}$	$\geq 5.1 \times 10^{20}$ [15]
$2\beta^+$	0ν	g.s.	$\geq 3.0 \times 10^{21} \text{ (CC 511 keV)}$	$\geq 1.2 \times 10^{21}$ [15]
$2\beta^+$	0ν	2+ 512	$\geq 2.5 \times 10^{21} \text{ (CC 511 keV)}$	$\geq 1.2 \times 10^{21} [15]$

efficiency of the setup to the $2\nu\varepsilon\beta^+$ decay (7.59%), the part of the energy spectrum in the energy interval (92.3%), the selection efficiency of the pulse-shape discrimination, time and energy cuts used to build the coincidence spectrum (98.4% in total), we have obtained the limit on the $2\nu\varepsilon\beta^+$ decay: $T_{1/2}^{2\nu\varepsilon\beta^+} \ge 2.0 \times 10^{21}$ yr. However, this limit depends on the model of background which has some uncertainty related to our limited knowledge of the radioactive impurities composition and localization in the setup. In addition, in some energy intervals (as in the considered above 250-1600 keV) the measured number of events is less than in the model of background. The authors of Ref. [42] suggest in such a situation to report the so-called sensitivity of the experiment defined as the average upper limit that would be obtained by an ensemble of experiments with the expected background and no true signal. Following these suggestions we took a more conservative, background-model-independent limit on the number of events of the effect searched for as $\lim S = 15.3$ counts, which leads to the following half-life limit on the $2\nu\varepsilon\beta^+$ decay of ¹⁰⁶Cd to the ground state of ¹⁰⁶Pd which we accept as a final result:

$$T_{1/2}^{2\nu\epsilon\beta^+} \ge 1.1 \times 10^{21} \text{yr}.$$

The excluded distribution of the $2\nu\varepsilon\beta^+$ decay of ¹⁰⁶Pd to the ground state of ¹⁰⁶Pd is presented in Fig. 9 together with the excluded spectra of the neutrinoless and two-neutrino doublepositron decay of ¹⁰⁶Cd. The limits on the processes with positron(s) emission for transitions to the ground and excited states of ¹⁰⁶Pd were obtained in a similar way mainly by using the coincidence of the ¹⁰⁶Cd WO₄ detector with annihilation γ quanta in the HPGe detectors (the limits are presented in Table I). A rather high sensitivity was achieved for the $\varepsilon\beta^+$ and $2\beta^+$ decay channels thanks to the comparatively high



FIG. 10. Energy spectrum of the ¹⁰⁶Cd WO₄ detector in coincidence with 616-keV γ quanta in the HPGe detector (filled histogram) acquired over 13 085 h. The excluded distributions of the $2\nu 2\varepsilon$ and $0\nu 2\varepsilon$ decay of ¹⁰⁶Cd to the 1128-keV excited level of ¹⁰⁶Pd with detection of 616-keV γ quanta in the HPGe detectors are shown.

probability to detect at least one annihilation γ quantum by the HPGe counters (e.g., 13.6% for the $2\beta^+$ decay).

Further suppression of the background was achieved by selection of events in the ¹⁰⁶Cd WO₄ detector in coincidence with the intense γ quanta in the HPGe counters expected in the double-electron capture in ¹⁰⁶Cd to the excited levels of ¹⁰⁶Pd. For instance, γ quanta with energy 616 keV expected in the 2ε decay of ¹⁰⁶Cd to the excited level 1128 keV of ¹⁰⁶Pd can be detected by the HPGe counters with a detection efficiency 2.42% (2.34%) in the two-neutrino (neutrinoless) process. As one can see in Fig. 10, there are only 17 (23) counts in the energy interval 50-700 keV (50-2500 keV). According to Ref. [42] one should take a sensitivity estimation of $\lim S =$ 8.4(9.7) counts. The energy intervals contain 78.2% (86.2%) of the simulated $2\nu 2\varepsilon$ ($0\nu 2\varepsilon$) spectra. Taking into account the efficiency of the pulse-shape discrimination, the time and energy coincidence with the γ quanta 616 keV (97.3% in total), the following half-life limit on the $2\nu 2\varepsilon$ ($0\nu 2\varepsilon$) decay of ¹⁰⁶Cd to the excited level 1128 keV of ¹⁰⁶Pd can be obtained: $T_{1/2}^{2\nu_{2}\epsilon} \ge 5.5 \times 10^{20} \text{ yr} (T_{1/2}^{0\nu_{2}\epsilon} \ge 5.1 \times 10^{20} \text{ yr})$. The excluded distributions of the $2\nu 2\varepsilon$ and $0\nu 2\varepsilon$ decays of 106 Cd to the 1128-keV excited level of ¹⁰⁶Pd are presented in Fig. 10.

Most of the limits on the double-electron capture in ¹⁰⁶Cd to the excited levels of ¹⁰⁶Pd were obtained in a similar way (including the transitions to the levels of ¹⁰⁶Pd with energies 2718 and 2748 keV where the resonant processes are possible). Typical detection efficiencies for the 2ε processes in ¹⁰⁶Cd to the excited levels of ¹⁰⁶Pd are 1.17–4.13% depending on the level and decay mode, while the number of counts varies from 83 to 0 depending on the energy in the HPGe detectors chosen to build the coincidence spectra. For example, 83 coincidence counts were obtained in the energy interval 50–2600 keV in CC with energy 512 keV in the HPGe counters, while 0 events were detected in the energy interval 650–2000 keV



FIG. 11. Part of the energy spectrum accumulated by the HPGe detector. Excluded peaks expected in the $0\nu 2K$, $0\nu LK$, and $0\nu 2L$ capture in ¹⁰⁶Cd to the ground state of ¹⁰⁶Pd are shown. Energies of the expected γ peaks are in keV.

in the coincidence with the energy 1160 keV in the HPGe detectors (resonant $0\nu 2K$ capture to the 2718-keV excited level of ¹⁰⁶Pd).

We have also used the data accumulated by the HPGe detectors to estimate limits on the 2β processes in ¹⁰⁶Cd. For instance, in neutrinoless 2ε capture we assume that the energy excess is taken away by bremsstrahlung γ quanta with energy $E_{\gamma} = Q_{2\beta} - E_{b1} - E_{b2} - E_{exc}$, where E_{bi} is the binding energy of *i*th captured electron on the atomic shell, and E_{exc} is the energy of the populated (g.s. or excited) level of ¹⁰⁶Pd. In case of transition to an excited level, in addition to the initial γ quantum, other γ 's will be emitted in the nuclear de-excitation process. For example, to derive a limit on the $0\nu 2K$ capture in ¹⁰⁶Cd to the ground state of ¹⁰⁶Pd the energy spectrum accumulated with the HPGe detectors was fitted in the energy interval 2700-2754 keV by a simple function (first-degree polynomial function to describe background plus Gaussian peak at the energy 2726.7 keV with the energy resolution FWHM = 4.4 keV to describe the effect searched for). The fit gives an area of the peak 6.2 ± 3.2 counts, with no evidence for the effect. According to Ref. [42] we took 11.4 events which can be excluded with 90% CL. Taking into account the detection efficiency for γ quanta with energy 2726.7 keV in the experimental conditions (1.89%) we have set the following limit for the $0\nu 2K$ capture of ¹⁰⁶Cd to the ground state of ¹⁰⁶Pd: $T_{1/2} \ge 4.2 \times 10^{20}$ yr. Limits on the $0\nu KL$ and $0\nu 2L$ capture in ¹⁰⁶Cd to the ground state of ¹⁰⁶Pd $(T_{1/2} \ge 1.3 \times 10^{21} \text{ yr and } T_{1/2} \ge 5.4 \times 10^{20} \text{ yr, respectively})$ were derived in the similar way. The excluded peaks are shown in Fig. 11. We would like to stress the certain advantages of isotope ¹⁰⁶Cd for which the high accuracy $Q_{2\beta}$ value is now available. Thanks to this feature one could distinguish clearly the 0v2K, 0vKL, and 0v2L modes of the decay using high-resolution HPGe detectors.

All the half-life limits obtained in the present work are summarized in Table I, where results of the most sensitive previous experiments are given for comparison.

1. Limit on ¹⁰⁶Cd $2v\varepsilon\beta^+$ decay matrix element

An upper limit on the effective nuclear matrix element for the $2\nu\varepsilon\beta^+$ decay $(M_{\text{eff}}^{2\nu\varepsilon\beta^+})$ can be obtained with the help of the following formula:

$$\lim M_{\rm eff}^{2\nu\varepsilon\beta^+} = 1/\sqrt{\lim T_{1/2}^{2\nu\varepsilon\beta^+}G^{2\nu\varepsilon\beta^+}},\tag{2}$$

where $G^{2\nu\epsilon\beta^+}$ is the phase space factor for the $2\nu\epsilon\beta^+$ decay, $M_{\rm eff}^{2\nu\epsilon\beta^+} = g_A^2(m_ec^2)M^{2\nu\epsilon\beta^+}$, g_A is the axial vector coupling constant, and m_ec^2 is the electron mass. Using the recently calculated phase space factors 702×10^{-24} yr⁻¹ [43] and 741×10^{-24} yr⁻¹ [44], we have obtained the following limit on the nuclear matrix element for the $2\nu\epsilon\beta^+$ decay of ¹⁰⁶Cd to the ground state of ¹⁰⁶Pd: $M_{\rm eff}^{2\nu\epsilon\beta^+} \leq 1.1$.

IV. CONCLUSIONS

An experiment to search for 2β decay of ¹⁰⁶Cd with enriched ¹⁰⁶Cd WO₄ crystal scintillator in coincidence with four crystals HPGe γ detector has been completed after 13085 h of data collection. New limits on 2ε , $\varepsilon\beta^+$, and $2\beta^+$ processes in ¹⁰⁶Cd were set on the level of $T_{1/2} >$ 10^{20} to 10^{21} yr. The highest sensitivity was achieved mainly by analysis of coincidence between the ¹⁰⁶Cd WO₄ and the HPGe detectors. For the decay channels with emission of positrons ($\varepsilon\beta^+$ and $2\beta^+$ decays) a higher sensitivity was achieved in coincidence with annihilation γ quanta, while coincidence of the ¹⁰⁶Cd WO₄ detector with intensive γ quanta in the HPGe counters gives the most stringent limits on

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the double electron capture in ¹⁰⁶Cd to excited levels of ¹⁰⁶Pd. The half-life limit on the two neutrino $\varepsilon\beta^+$ decay, $T_{1/2} \ge 1.1 \times 10^{21}$ yr, reached the region of some theoretical predictions [19,46–50]. By using the half-life limit the nuclear matrix element for the $2\nu\varepsilon\beta^+$ decay of ¹⁰⁶Cd to the ground state of ¹⁰⁶Pd can be bounded as $M_{\text{eff}}^{2\nu\varepsilon\beta^+} \le 1.1$. The resonant neutrinoless double-electron captures to the 2718-, 2741-, and 2748-keV excited states of ¹⁰⁶Pd are restricted on the level of $T_{1/2} \ge (8.5 \times 10^{20} \text{ to } 1.4 \times 10^{21})$ yr. Unfortunately, the experiment has no competitive sensitivity to the most probable $2\nu 2K$ channel of the decay due to the high activity of ¹¹³Cd^m in the ¹⁰⁶Cd WO₄ crystal scintillator. Advancement of the experiment is in progress using the ¹⁰⁶Cd WO₄ scintillation detectors in close geometry to improve the detection efficiency to γ quanta emitted in the double- β processes in ¹⁰⁶Cd.

ACKNOWLEDGMENTS

The authors from the Institute for Nuclear Research (Kyiv, Ukraine) were supported in part by Project No. CO-1-2/2015 of the program of collaboration with the Joint Institute for Nuclear Research (Dubna) "Promising Basic Research on High Energy and Nuclear Physics" for 2014–2015 of the National Academy of Sciences of Ukraine. We are grateful to S. R. Elliott for his note on presence of ¹¹⁶Cd $2\nu 2\beta^{-}$ decay in ¹⁰⁶Cd WO₄ data. We thank Prof. H. Ejiri for useful discussions.

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