New results of ^{116}Cd double β decay study with $^{116}CdWO_4$ scintillators

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A new phase of a ^{116}Cd double β decay experiment is in progress in the Solotvina Underground Laboratory. Four enriched $^{116}CdWO_4$ scintillators with total mass of 339 g are used in a setup, whose active shield is made of 15 natural $CdWO_4$ crystals (20.6 kg). The background rate in the energy interval 2.5–3.2 MeV is 0.03 counts/yr kg keV. The half-life for $2\nu2\beta$ decay of ^{116}Cd is measured as $T_{1/2}(2\nu)=2.6\pm0.1(\mathrm{stat})^{+0.7}_{-0.4}(\mathrm{syst})\times10^{19}$ yr. The $T_{1/2}$ limits for neutrinoless 2β decay of ^{116}Cd are set at $T_{1/2}\geqslant0.7(2.5)\times10^{23}$ yr at 90%(68%) C.L. for transition to ground state of ^{116}Sn, while for decays to the first 2^+_1 and second 0^+_1 excited levels of ^{116}Sn at $T_{1/2}\geqslant1.3(4.8)\times10^{22}$ yr and $\geqslant0.7(2.4)\times10^{22}$ yr with 90%(68%) C.L., respectively. For $0\nu2\beta$ decay with emission of one or two Majorons, the limits are $T_{1/2}(0\nu M1)\geqslant3.7(5.8)\times10^{21}$ yr and $T_{1/2}(0\nu M2)\geqslant5.9(9.4)\times10^{20}$ yr at 90%(68%) C.L. Restrictions on the value of the neutrino mass, right-handed admixtures in the weak interaction, and the neutrino-Majoron coupling constant are derived as $m_\nu\leqslant2.6(1.4)$ eV, $\eta\leqslant3.9\times10^{-8}$, $\lambda\leqslant3.4\times10^{-6}$, and $g_M\leqslant12(9.5)\times10^{-5}$ at 90%(68%) C.L., respectively.

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I. INTRODUCTION

Neutrinoless (0ν) double β decay is forbidden in the standard model (SM) since it violates lepton number (L) conservation. However, many extensions of the SM incorporate L violating interactions and thus could lead to the $0\nu2\beta$ decay [1,2]. Currently, in addition to conventional neutrino (ν) exchange mechanism, there are many other possibilities to trigger this process [2]. In that sense neutrinoless 2β decay has a great conceptual importance due to the strong Schechter-Valle theorem [3] obtained in a gauge theory of the weak interaction that a nonvanishing $0\nu2\beta$ decay rate requires neutrino to be massive Majorana particle, independent of which mechanism induces it. Therefore, at present $0\nu2\beta$ decay is considered as a very powerful test of new physical effects beyond the SM, and even the absence of this process—at the present level of sensitivity—would help to restrict or narrow this wide choice of theoretical models. At the same time $0\nu2\beta$ decay is very important in the light of the measured deficit of the atmospheric neutrinos flux [4,5] and the result of the LSND accelerator experiment [4,6], which could be explained by means of the neutrino oscillations requiring in turn nonzero neutrino masses (m_ν). However, oscillation experiments are sensitive to neutrino mass difference, while measured $0\nu2\beta$ decay rate can give the absolute value of the m_ν scale, and hence provide a crucial test of m_ν models.

Despite numerous attempts to observe $0\nu2\beta$ decay from 1948 up to the present [1] this process still remains unobserved. The highest $T_{1/2}(0\nu)$ limits were set in direct experiments with several nuclides: $T_{1/2}\geqslant10^{22}$ yr for ^{82}Se [7], ^{100}Mo [8], ^{116}Cd [9]; $T_{1/2}\geqslant10^{23}$ yr for ^{130}Te [10] and ^{136}Xe [11]; and $T_{1/2}\geqslant10^{25}$ yr for ^{76}Ge [12,13].

With the aim to enlarge the number of 2β decay candidate nuclides studied at a sensitivity comparable with that for ^{76}Ge and ^{136}Xe (neutrino mass limit of 0.5–2 eV), cadmium tungstate crystal scintillators, enriched in ^{116}Cd to 83%, were developed and exploited in ^{116}Cd research [14,9]. The measurements were carried out in the Solotvina Underground Laboratory in a salt mine 430 m underground ($\simeq$1000 m w. e.) [15]. In the first phase of the experiment only one $^{116}CdWO_4$ crystal (15.2 cm^3) was used. It was viewed by a photomultiplier tube (PMT) through a light guide 51 cm long and placed inside a plastic scintillator ($\oslash38\times115$ cm) which served as a veto counter. A passive shield of high purity (HP) copper (5 cm), lead (23 cm), and polyethylene (16 cm) surrounded the plastic counter. The background rate in the energy range 2.7–2.9 MeV ($Q_{2\beta}=2805$ keV [16]) was equal $\approx$0.6 counts/yr kg keV. With 19175 h statistics the half-life limit for $0\nu2\beta$ decay of ^{116}Cd was set as $T_{1/2}(0\nu)\geqslant3.2\times10^{22}$ yr (90% C.L.) [9], which corresponds to the restriction on the neutrino mass $m_\nu\leqslant3.9$ eV [17]. Limits on $0\nu2\beta$ decay with emission of one ($M1$) or two ($M2$) Majorons were obtained too: $T_{1/2}(0\nu M1)\geqslant1.2\times10^{21}$ yr and $T_{1/2}(0\nu M2)\geqslant2.6\times10^{20}$ yr (90% C.L.) [18]. In the present paper new and advanced results of ^{116}Cd research obtained with the help of an upgraded apparatus are described.

II. NEW SETUP WITH FOUR $^{116}CdWO_4$ DETECTORS

A. Setup and measurements

In order to enhance the sensitivity of the ^{116}Cd experiment, the following improvements were scheduled: increase of the number of ^{116}Cd nuclei, reduction of the background

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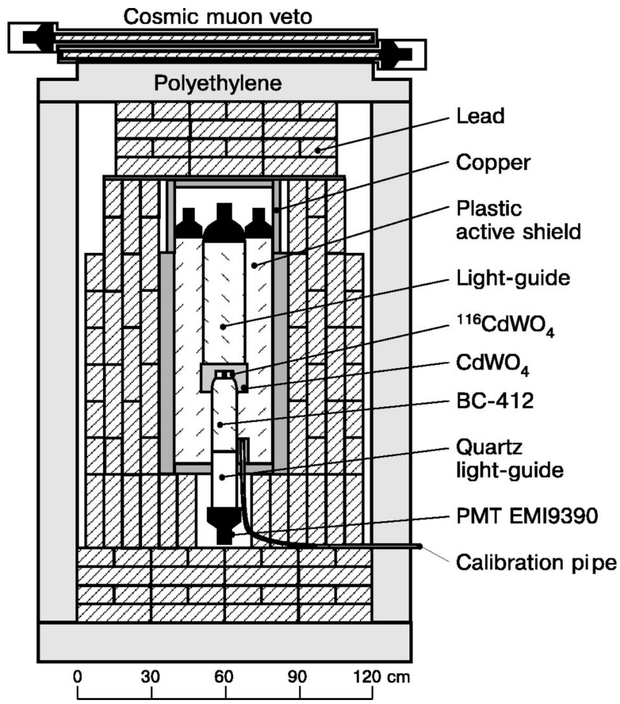

FIG. 1. The scheme of the new setup with four $^{116}CdWO_4$ detectors.

and improvement of the data taking and processing [9]. With this aim the new setup with four enriched $^{116}CdWO_4$ crystals (total mass 339 g) has been mounted in the Solotvina Laboratory in August 1998. All materials used in the installation were previously tested and selected for low radioactive impurities in order to reduce their contributions to background.

In the new apparatus, a scheme of which is shown in Fig. 1, four enriched crystals are viewed by the PMT (EMI9390) through one light-guide 10 cm in diameter and 55 cm long, which is composed of two glued parts: quartz 25 cm long and plastic scintillator (Bicron BC-412) 30 cm long. The $^{116}CdWO_4$ crystals are surrounded by an active shield made of 15 natural $CdWO_4$ scintillators of large volume ($\simeq 200$ cm^3 each) with total mass of 20.6 kg. Due to the high purity of the $CdWO_4$ crystals [19] and their large density (≈ 8 g/cm^3) this active shield reduces background effectively. The veto crystals are viewed—by a low background PMT (⌀17 cm)—through an active plastic light guide (⌀17×49 cm). In turn the whole array of $CdWO_4$ counters is placed inside an additional active shield made of polystyrene-based plastic scintillator with dimensions 40×40×95 cm. Thus, together with both active light guides (connected with enriched and natural crystals on opposite sides), a complete 4π active shield of the main $^{116}CdWO_4$ detectors is provided.

The outer passive shield consists of HP copper (thickness 3–6 cm), lead (22.5–30 cm), and polyethylene (16 cm). Two plastic scintillators (120×130×3 cm) are installed above the passive shield to provide a cosmic muons veto. Because air in the Solotvina Laboratory can be contaminated by radon (at the level $\leqslant 30$ Bq/m^3) the setup is isolated carefully against air penetration. All cavities inside the shield are filled up by pieces of plexiglass, and HP Cu shield is sealed with the help of silicon glue and enclosed inside a tight mylar envelope.

The new event-by-event data acquisition is based on two IBM personal computers (PC) and a CAMAC crate with electronic units. For each event the following information is stored on the hard disc of the first computer. The amplitude (energy) of a signal, its arrival time and the following additional tags: the coincidence between different detectors; the signal of radio-noise detection system; triggers for light emitting diode (LED); and pulse shape digitizer. The second computer records the pulse shape of the $^{116}CdWO_4$ scintillators in the energy range 0.25–5 MeV. This complementary system is developed on the basis of a fast 12 bit ADC (Analog Devices AD9022) and is connected with computer by parallel digital I/O board (PC-DIO-24 from National Instruments) [22]. Two PC-DIO-24 boards are used to link both computers and establish—with the help of proper software—a one-to-one correspondence between the pulse shape data recorded by the second computer and the information stored in the first PC.

The energy scale and resolution of the main detector—four enriched crystals taken as a whole—were determined in the measurements with different sources (^{22}Na, ^{40}K, ^{60}Co, ^{137}Cs, ^{207}Bi, ^{226}Ra, ^{232}Th, and ^{241}Am). The energy dependence of the resolution can be expressed (for the energy above 50 keV) as FWHM (keV) $=\sqrt{-226+16.6E+6.42\times10^{-3}E^2}$, where energy E is in keV. For instance, the resolution (FWHM) was equal to 14.5% at 1064 keV and 11% at 2615 keV. The full energy peaks are well fitted in the energy region 0.06–2.6 MeV by a Gaussian function with typical value $\chi^2=0.8$–1.9. Moreover, the calibration spectra of the ^{232}Th source were simulated with the help of GEANT3.21 package [23] and event generator DECAY4 [24] (the last defines initial kinematics of the events). The simulated ^{232}Th spectra are in good agreement with the measured ones confirming the assumption of a Gaussian peak shape. In particular for the 2615 keV peak of ^{208}Tl—which is close to the 2β decay energy of ^{116}Cd—the value of $\chi^2=1.3$.

Also, the relative light yield for α particles as compared with that for electrons (α/β ratio) and energy resolution were measured with α sources and corrected by using time-amplitude analysis (see below) as the following: $\alpha/\beta=0.15(1)+7\times10^{-6}E_\alpha$ and $\mathrm{FWHM}_\alpha(\mathrm{keV})=0.053E_\alpha$ (E_α is in keV). The routine calibration is carried out weekly with a ^{207}Bi source (570, 1064, and 1770 keV) and once per two weeks with ^{232}Th (2615 keV). The dead time of the spectrometer and data acquisition is monitored permanently with the help of an LED optically connected with the main PMT. The actual dead time value is ≈4.2% (≈3% is owing to random coincidence between the main and shield detectors; ≈1.2% is caused by miscounts of the data acquisition).

The background spectrum measured during 4629 h in the new installation with four $^{116}CdWO_4$ crystals is given in Fig. 2, where the old data obtained with one $^{116}CdWO_4$ crystal of 121 g are also shown for comparison. As is visible from this figure, the background is decreased in the whole energy range, except for the β spectrum of ^{113}Cd ($Q_\beta=316$ keV), whose abundance in $^{116}CdWO_4$ crystals is ≈2% [14]. In the energy region 2.5–3.2 MeV—where the peak of $0\nu2\beta$ decay

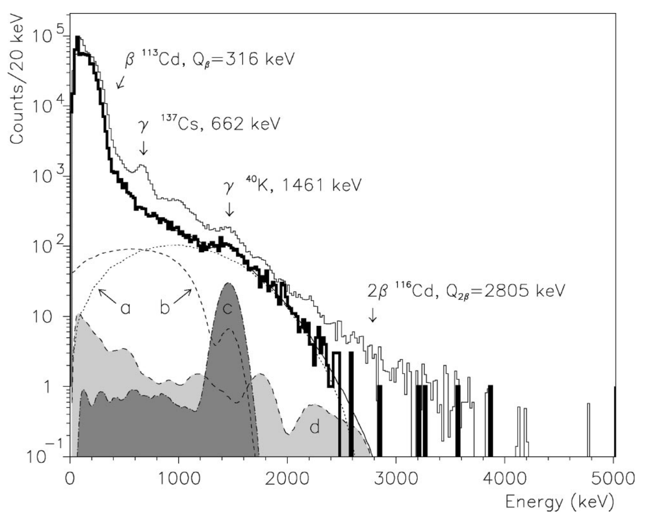

FIG. 2. Background spectrum of ^{116}CdWO$_4$ detectors (339 g) measured in the setup with four enriched crystals during 4629 h (solid histogram). The old data obtained with one ^{116}CdWO$_4$ crystal (121 g; 19986 h) is shown for comparison (thin histogram; the data are normalized to 4629 h and mass of the new detector). The background components used for fit in the energy region 900–2900 keV: (a) $2\nu 2\beta$ decay of ^{116}Cd [fit value is $T_{1/2}(2\nu) = 2.6(1)\times 10^{19}$ yr]; (b) ^{40}K inside the ^{116}CdWO$_4$ detector [activity value from the fit is 0.8(2) mBq/kg]; (c) ^{40}K in the shielding CdWO$_4$ crystals [fit value is 2.1(3) mBq/kg]; (d) ^{226}Ra and ^{232}Th contamination of PMTs.

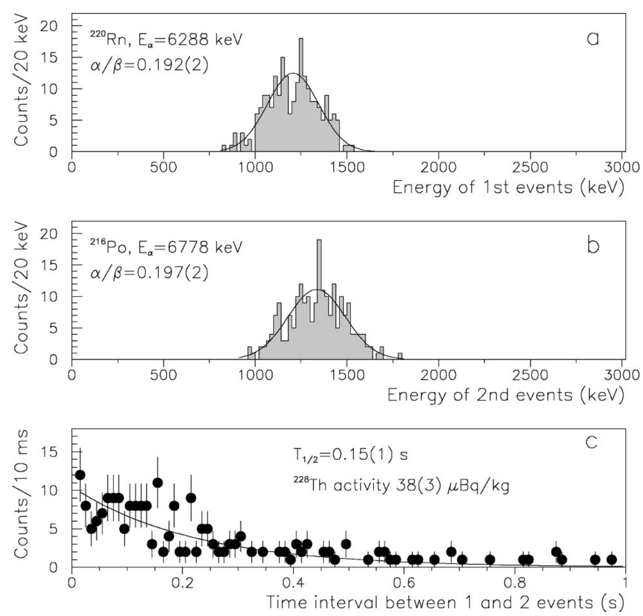

FIG. 3. The energy spectra of the first (a) and second (b) α particles from the ^{220}Rn$\rightarrow$^{216}Po$\rightarrow$^{212}Pb chain selected by time-amplitude analysis from ^{116}CdWO$_4$ data. Their equivalent energies in the β/γ energy scale are near 5 times smaller because the relative light yield for α particles as compared with that for electrons (α/β ratio) is ≈ 0.2. (c) Time distribution between the first and second events together with exponential fit [$T_{1/2} = 0.15(1)$ s, while the table value is $T_{1/2} = 0.145(2)$ s [25]].

of ^{116}Cd is expected—the background rate is reduced to a value of 0.03 counts/yr kg keV (only four events in the energy window 2.5–3.2 MeV were detected during 4629 h), 20 times lower than in the previous setup. It is achieved, first, due to improvement of passive and active shield, and secondly, as a result of data processing advance (time-amplitude and pulse-shape analysis), which are described below.

B. Time-amplitude analysis of the data

The energy and arrival time of each event can be used for analysis and selection of some decay chains in ^{232}Th, ^{235}U, and ^{238}U families (see, e.g., Refs. [11,14]). As an example (important in the following for the background rejection in the energy range of $0\nu 2\beta$ decay), we consider here in detail the time-amplitude analysis of the following sequence of α decays from ^{232}Th family: ^{220}Rn ($Q_\alpha = 6.40$ MeV, $T_{1/2} = 55.6$ s)$\rightarrow$^{216}Po ($Q_\alpha = 6.91$ MeV, $T_{1/2} = 0.145$ s)$\rightarrow$^{212}Pb. Because the energy of α particles from ^{220}Rn decay corresponds to $\simeq 1.2$ MeV in β/γ scale of ^{116}CdWO$_4$ detector, the events in the energy region 0.7–1.8 MeV were used as triggers. Then all events (within 0.9–1.9 MeV) following the triggers in the time interval 10–1000 ms (containing 94.5% of ^{216}Po decays) were selected. The spectra of the ^{220}Rn and ^{216}Po α decays obtained in this way from data—as well as the distribution of the time intervals between the first and second events—are presented in Fig. 3. It is evident from this figure that the selected spectra and time distribution are in an excellent agreement with those expected from α particles of ^{220}Rn and ^{216}Po. Using these results and taking into account the efficiency of the time-amplitude analysis and the number of accidental coincidences (three pairs from 218 selected), the determined activity of ^{228}Th (^{232}Th family) inside the ^{116}CdWO$_4$ crystals is as low as 38(3) μBq/kg.

The same technique was applied to the sequence of α decays from the ^{235}U family: ^{223}Ra ($Q_\alpha = 5.98$ MeV, $T_{1/2} = 11.44$ d)$\rightarrow$^{219}Rn ($Q_\alpha = 6.95$ MeV, $T_{1/2} = 3.96$ s)$\rightarrow$^{215}Po ($Q_\alpha = 7.53$ MeV, $T_{1/2} = 1.78$ ms)$\rightarrow$^{211}Pb. For the fast couple (^{219}Rn$\rightarrow$^{215}Po) all events within 0.8–1.8 MeV were used as triggers, while a time interval 1–10 ms (65.7% of ^{215}Po decays) and an energy window 0.9–2.0 MeV were set for the second events. The obtained α peaks correspond to an activity of 5.5(14) μBq/kg for the ^{227}Ac impurity in the crystals.

As regards the ^{226}Ra chain (^{238}U family) the following sequence of β and α decays was analyzed: ^{214}Bi ($Q_\beta = 3.27$ MeV, $T_{1/2} = 19.9$ m)$\rightarrow$^{214}Po ($Q_\alpha = 7.83$ MeV, $T_{1/2} = 164.3$ μs)$\rightarrow$^{210}Pb. For the first and second events the energy threshold was equal 0.1 MeV, and a time interval of 100–1000 μs (64.1% of ^{214}Po decays) was used. While the obtained spectrum of the first pulses agrees with the model of the β decay of ^{214}Bi, and the distribution of the time intervals between the first and second events can be fitted by an exponent with $T_{1/2} = 140^{+30}_{-20}$ μs (in reasonable agreement with the ^{214}Po half-life value), the spectrum of the second events is continuous, contrary to the anticipated α peak of ^{214}Po. Part of this continuous distribution may be explained by ^{226}Ra contamination of the materials neighboring the ^{116}CdWO$_4$ crystals (optical grease, Teflon, Mylar, radon in air), while another part is caused by ^{226}Ra decays in the crystals. Under such an assumption activity limits for ^{226}Ra contaminations are derived as $\leqslant 0.13(3)$ Bq/kg for optical grease, $\leqslant 8$ mBq/kg for Teflon, $\leqslant 1.8$ μBq/dm^2 for Mylar, and $\leqslant 5$ μBq/kg for $^{116}CdWO_4$, whose values do not contradict bounds obtained earlier [18]. To prove these assumptions, the events belonging to the $^{214}Bi \rightarrow ^{214}Po \rightarrow ^{210}Pb$ chain were independently searched for in the time window of 5–88 μs (28.9% of ^{214}Po decays) with the help of pulse shape analysis (see below). For both events the energy threshold was ≈ 0.3 MeV. The result obtained (^{226}Ra activity in the $^{116}CdWO_4$ crystals $\leqslant 14$ μBq/kg) is similar to that of the time-amplitude analysis. Finally, all couples of events found for ^{232}Th, ^{235}U, and ^{238}U families as described above were eliminated from the measured data.

C. Pulse-shape discrimination

The pulse shape of the $^{116}CdWO_4$ scintillators in the energy region of 0.25–5 MeV is digitized by a 12 bit ADC and stored in 2048 channels with 50 ns channel's width. Due to different shapes of scintillation signal for various kinds of sources[1] (α particles, protons, γ quanta and cosmic muons were investigated), the pulse-shape (PS) discrimination method based on the optimal digital filter [20] was developed and clear discrimination between γ rays (electrons) and α particles was achieved [22].

The pulse shapes of enriched crystals were measured for α particles with an ^{241}Am source and for γ rays with ^{60}Co, ^{137}Cs, ^{207}Bi, and ^{232}Th sources in the special calibration runs.[2] To provide an analytic description of the α or γ signals $f_\alpha(t)$ and $f_\gamma(t)$ the pulse shape resulting from the average of a large number of individual events has been fitted with the sum of three (for α particles) or two (for γ's) exponents, giving the reference pulse shapes $\overline{f_\alpha}(t)$ and $\overline{f_\gamma}(t)$ (see for more details Ref. [22]). In the data processing the digital filter is applied to each experimental signal $f(t)$ with aim to obtain the numerical characteristic of its shape [shape indicator (SI)] defined as $\mathrm{SI}=\Sigma_k f(t_k)\cdot P(t_k-t_0)$, where the sum is over all time bins (from $k=1$ to $k=2048$), $f(t_k)$ is the digitized amplitude of a given signal (normalized to its area) at the time t_k. The weight function $P(t_k-t_0)$ is determined as $P(t)=\{\overline{f_\alpha}(t)-\overline{f_\gamma}(t)\}/\{\overline{f_\alpha}(t)+\overline{f_\gamma}(t)\}$, and t_0 is the time origin of the signal. The measured with sources SI distributions are well described by a Gaussian functions,

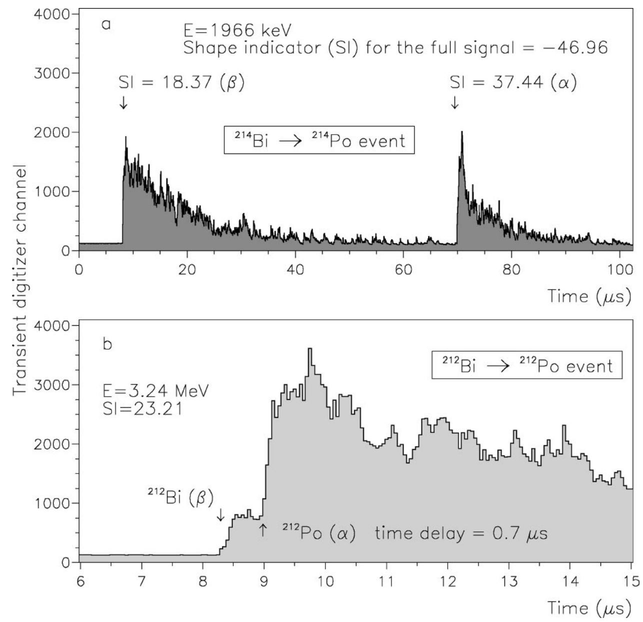

FIG. 4. (a) Example of a double pulse with total energy 1.97 MeV. The shape indicators for the full signal and separately for its first and second parts are $\mathrm{SI}_\Sigma=-47$; $\mathrm{SI}_1=18.4$ (close to SI_γ) and $\mathrm{SI}_2=37.4$ (close to SI_α). Most probably, this is the couple of successive decays ^{214}Bi (β)$\rightarrow^{214}$Po ($\alpha; T_{1/2}=164.3$ μs)$\rightarrow^{210}$Pb. (b) Probable event of the chain ^{212}Bi (β)$\rightarrow^{212}$Po ($\alpha; T_{1/2}=0.3$ μs) $\rightarrow^{208}$Pb.

whose mean values and standard deviations σ_α and σ_γ have a slight energy dependence.[3] For 0.9 MeV γ quanta $\mathrm{SI}_\gamma=18\pm3$, while for 4.8 MeV α particles $\mathrm{SI}_\alpha=29.0\pm3.6$. It allows us to determine the efficiency of the PS event selection for the different chosen intervals of SI values ($\pm\sigma$, $\pm2\sigma$, etc.).

The PS selection technique ensures the very important possibility to discriminate ''illegal'' events: double pulses, α events, etc., and thus to suppress background. An example of a double pulse is shown in Fig. 4(a). Value of the shape indicator for the full signal is $\mathrm{SI}=-47$; for the first pulse $\mathrm{SI}_1=18.4$ (hence it corresponds to γ or β particle), for the second pulse $\mathrm{SI}_2=37.4$ (α particle). The energy release is 1.97 MeV, and without PS analysis it would be a candidate event for $2\nu2\beta$ decay of ^{116}Cd.

Since the shape indicator characterizes the full signal, it is also useful to examine the pulse front edge. For example, it was found that at least 99% of ''pure'' γ events (measured with calibration ^{232}Th source) satisfy the following restriction on pulse rise time: $\Delta t(\mu\mathrm{s})\leqslant 1.24-0.5E_\gamma+0.078E_\gamma^2$, where E_γ is dimensionless variable expressed in MeV. Hence, this filter was applied to the background data, and all

[1]It is known that scintillation efficiency and pulse shape of inorganic crystals depend on the local density of the energy released, hence allowing to identify the incoming radiation (see, e.g., Refs. [20,21]).

[2]Because γ rays interact with matter by mean of the energy transfer to electrons, it was assumed that pulse shapes for electrons and γ's are the same. This statement was proved in the measurement with conversion electrons of ^{207}Bi by using the signal of the thin plastic scintillator (placed between source and detector) as signature of the electron hitting the $^{116}CdWO_4$ crystal.

[3]For the γ's (300–3200 keV) $\mathrm{SI}_\gamma=18.09-(4.5\times10^{-5}E_\gamma)$, $\sigma_\gamma=2.61-(4.7\times10^{-4}E_\gamma)+707/E_\gamma$, while for the α particles (4000–6000 keV) $\mathrm{SI}_\alpha=29.0$; $\sigma_\alpha=5.11-(5.52\times10^{-4}E_\alpha)+5520/E_\alpha$. Here all variables are dimensionless (E_γ and E_α are expressed in keV).

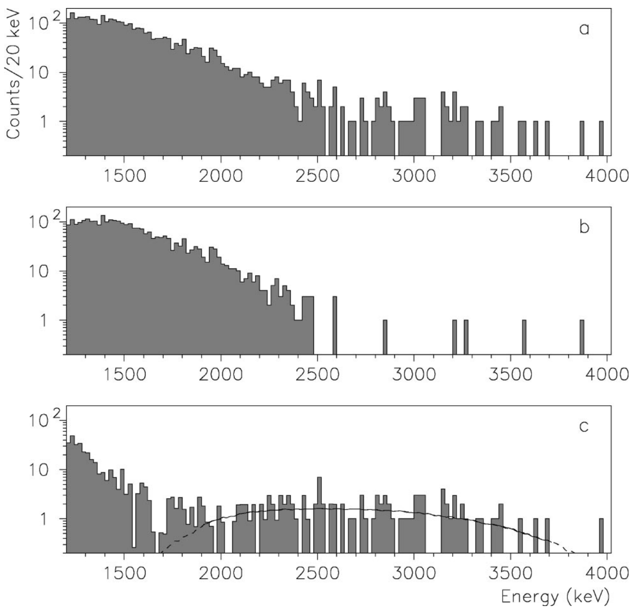

FIG. 5. (a) Initial spectrum of $^{116}CdWO_4$ crystals (339 g, 4629 h) in anticoincidence with shielding detectors without pulse-shape discrimination; (b) PS selected β/γ events (see text); (c) the difference between spectra in (a) and (b) together with the fit by the response function for ^{212}Bi→^{212}Po→^{208}Pb decay chain. The fit value is 37(4) μBq/kg for ^{228}Th activity inside $^{116}CdWO_4$ crystals.

events, which do not pass the test, were excluded from the residual β/γ spectrum.

The results of PS analysis of the data are presented in Fig. 5. The initial (without PS selection) spectrum of the $^{116}CdWO_4$ scintillators in the energy region 1.2–4 MeV—collected during 4629 h in anticoincidence with active shield—is depicted in Fig. 5(a), while the spectrum after PS selection of the β/γ events, whose SI lie in the interval $\mathrm{SI}_\gamma - 3.0\sigma_\gamma \leqslant \mathrm{SI} \leqslant \mathrm{SI}_\gamma + 2.4\sigma_\gamma$ and $\Delta t(\mu\mathrm{s}) \leqslant 1.24 - 0.5E_\gamma + 0.078E_\gamma^2$ (98% of β/γ events), is shown in Fig. 5(b). From these figures the background reduction due to pulse-shape analysis is evident. Further, Fig. 5(c) represents the difference between spectra in Figs. 5(a) and 5(b). These events, at least for the energy above 2 MeV, can be produced by ^{228}Th activity from the intrinsic contamination of the $^{116}CdWO_4$ crystals (measured by the time-amplitude analysis as described above). Indeed, two decays in the fast chain ^{212}Bi ($Q_\beta = 2.25$ MeV)→^{212}Po ($Q_\alpha = 8.95$ MeV, $T_{1/2} = 0.3$ μs)→^{208}Pb cannot be time resolved in the $CdWO_4$ scintillator (with an exponential decay time $\simeq 15$ μs [22]) and will result in one event. The example of such an event—recorded by the PS acquisition system—is depicted in Fig. 4(b). To determine the residual activity of ^{228}Th in the crystals, the response function of $^{116}CdWO_4$ detectors for the ^{212}Bi→^{212}Po→^{208}Pb chain was simulated with the help of GEANT3.21 code and event generator DECAY4. The simulated function is shown in Fig. 5(c), from which one can see that the high-energy part of the experimental spectrum is well reproduced ($\chi^2 = 1.3$) by the expected response for ^{212}Bi →^{212}Po→^{208}Pb decays.[4] Corresponding activity of ^{228}Th inside the $^{116}CdWO_4$ crystals, deduced from the fit in the 1.9–3.7 MeV energy region, is 37(4) μBq/kg, that is in a good agreement with the value determined by the time-amplitude analysis of the chain ^{220}Rn→^{216}Po→^{212}Pb. In addition, the front edge analysis of 80 events with the energy 2.0–4.2 MeV ($\mathrm{SI} \geqslant \mathrm{SI}_\gamma + 2.54\sigma_\gamma$; $\Delta t \geqslant 0.2$ μs) was fulfilled and the half-life derived from the average time delay between the first and second part of the signal [see Fig. 4(b)] is $T_{1/2} = 0.31(6)$ μs, in agreement with the ^{212}Po table value $T_{1/2} = 0.299(2)$ μs [25].

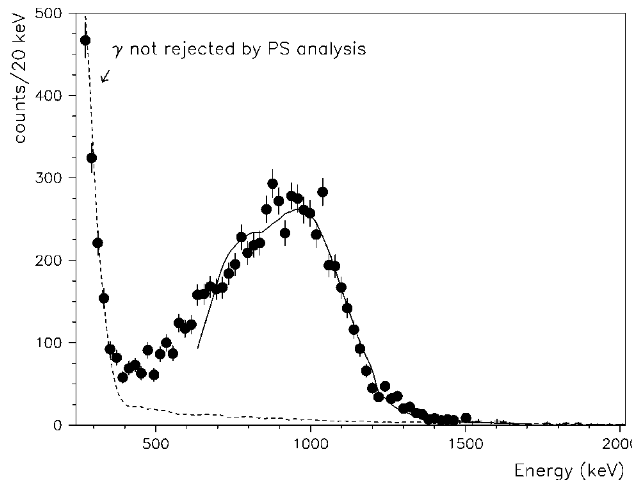

FIG. 6. Spectrum after PS selection of the background events, whose SI lies in the interval $\mathrm{SI}_\gamma + 2.4\sigma_\gamma < \mathrm{SI} < \mathrm{SI}_\alpha + 2.4\sigma_\alpha$ (it contains ≈90% of all α events). The model distribution (smooth line) includes all α particles from chains in ^{232}Th and ^{238}U families. The total α activity of the $^{116}CdWO_4$ crystals is derived as 1.4(3) mBq/kg.

Figure 6 represents the spectrum after PS selection of the background events, whose SI lie in the interval $\mathrm{SI}_\gamma + 2.4\sigma_\gamma < \mathrm{SI} < \mathrm{SI}_\alpha + 2.4\sigma_\alpha$ (≈90% of α events). The obtained distribution with maximum at 0.95 MeV is well reproduced by the model, which includes all α particles from chains in ^{232}Th and ^{238}U families. The total α activity of the $^{116}CdWO_4$ crystals deduced from Fig. 6 is 1.4(3) mBq/kg. This value can be adjusted with the activities determined by the time-amplitude analysis under usual (for crystals) assumption that secular radioactive equilibriums in some chains of ^{232}Th and ^{238}U families (such as ^{230}Th→^{226}Ra chain) are broken.

III. RESULTS AND DISCUSSION

A. Two-neutrino double β decay of ^{116}Cd

To determine the half-life of two-neutrino 2β decay of ^{116}Cd, the background in the energy interval 900–2900 keV was simulated with the help of GEANT3.21 package and event generator DECAY4. In addition to ^{116}Cd two neutrino 2β decay distribution, only three components shown in Fig. 2 were

[4]The rest of spectrum below 1.9 MeV [Fig. 5(c)] can be explained as high-energy tail of the PS selected α particles (see Fig. 6).

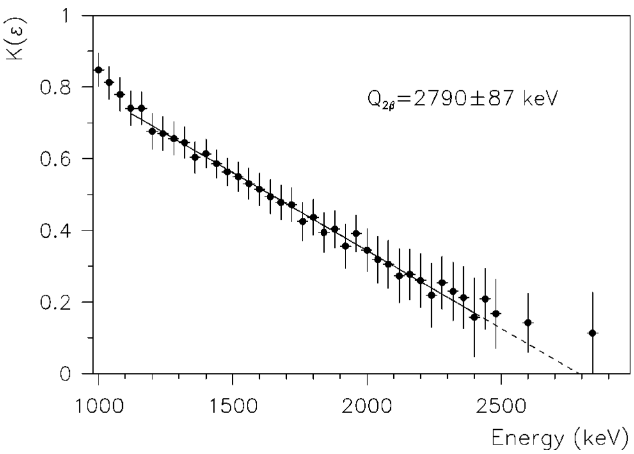

FIG. 7. The $2\nu2\beta$ decay Kurie plot and its fit by the straight line in 1100–2400 keV region.

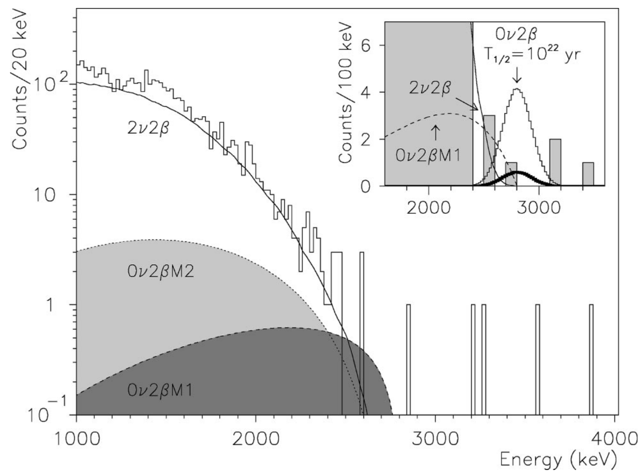

FIG. 8. Part of the experimental spectrum of the $^{116}CdWO_4$ detectors measured during 4629 h (histogram) together with the fit from $2\nu2\beta$ contribution ($T_{1/2}=2.6\times10^{19}$ yr). The smooth curves $0\nu2\beta M1$ and $0\nu2\beta M2$ are excluded with 90% C.L. distributions of $0\nu M1$ and $0\nu M2$ decay of ^{116}Cd with $T_{1/2}=3.7\times10^{21}$ yr and $T_{1/2}=5.9\times10^{20}$ yr, respectively. In the insert the expected peak from $0\nu2\beta$ decay with $T_{1/2}(0\nu)=1.0\times10^{22}$ yr is shown together with the excluded (90% C.L.) distribution (solid histogram) with $T_{1/2}(0\nu)=7.0\times10^{22}$ yr.

used to build up the background model: ^{40}K contamination of the enriched and natural $CdWO_4$ scintillators, whose activity limits of less than 4 mBq/kg were established earlier [19], and external γ background caused by ^{232}Th and ^{238}U contamination of the PMTs (one PMT for $^{116}CdWO_4$ crystals; one for $CdWO_4$; two for plastic active shield).[5] This simple background model describes experimental data in the chosen energy interval 900–2900 keV reasonably well ($\chi^2=1.3$) and gives the following results: the activities of ^{40}K inside the enriched and natural $CdWO_4$ crystals are equal 0.8(2) and 2.1(3) mBq/kg, respectively; the half-life of two-neutrino 2β decay of ^{116}Cd is $T_{1/2}(2\nu)=2.6(1)\times10^{19}$ yr (only statistical uncertainties are given, while systematical errors are pointed below).

Taking advantage of the high statistics in our experiment (approximately 3600 events of ^{116}Cd two neutrino 2β decay are contained within the interval 900–2900 keV), we can prove our model with the help of experimental $2\nu2\beta$ decay Kurie plot: $K(\varepsilon)=[S(\varepsilon)/\{(\varepsilon^4+10\varepsilon^3+40\varepsilon^2+60\varepsilon+30)\varepsilon\}]^{1/5}$, where S is the number of events with the energy ε (in electron mass units) in the experimental spectrum after background subtraction. For the real $2\nu2\beta$ decay events such a Kurie plot should be the straight line $K(\varepsilon)\sim(Q_{2\beta}-\varepsilon)$, where $Q_{2\beta}$ is the 2β energy release. From Fig. 7, where our experimental Kurie plot is depicted, one can see that in the region 1.1–2.4 MeV it is well fitted by the straight line with $Q_{2\beta}=2790(87)$ keV [the table value is $Q_{2\beta}=2805(4)$ keV]. To take into account the energy resolution of the detector the experimental spectrum was also fitted by the convolution of the theoretical $2\nu2\beta$ distribution $\rho(\varepsilon)=A\cdot\varepsilon(\varepsilon^4+10\varepsilon^3+40\varepsilon^2+60\varepsilon+30)\cdot(Q_{2\beta}-\varepsilon)^5$ with the detector resolution function (Gaussian with FWHM determined as described above) with A and $Q_{2\beta}$ values as free parameters. This fit in the energy region 1.2–2.8 MeV yields a very similar value $Q_{2\beta}=2779(52)$ keV and an A corresponding to $T_{1/2}(2\nu)=2.5(3)\times10^{19}$ yr, thus justifying our assumption that experimental data in the region above 1.2 MeV are related mainly with ^{116}Cd two neutrino 2β decay. In fact, a signal to background ratio deduced from our data is 4:1 for the energy interval 1.2–2.9 MeV, and 15:1 for the energy range 1.6–2.9 MeV, which are higher than those reached up-to-date in other 2β decay experiments [1,2].

To estimate systematical uncertainties of the measured half-life, different origins of errors were taken into account, whose contributions are listed in Table I. The final value is equal to

$$T_{1/2}(2\nu)=2.6\pm0.1(\mathrm{stat})^{+0.7}_{-0.4}(\mathrm{syst})\times10^{19}\ \mathrm{yr}.$$

Our result is in agreement with those measured earlier ($T_{1/2}(2\nu)=2.6^{+0.9}_{-0.5}\times10^{19}$ yr [26] and $T_{1/2}(2\nu)=2.7^{+0.5}_{-0.4}(\mathrm{stat})^{+0.9}_{-0.6}(\mathrm{syst})\times10^{19}$ yr [14]) and disagrees to some extent with the value $T_{1/2}(2\nu)=3.75\pm0.35(\mathrm{stat})\pm0.21(\mathrm{syst})\times10^{19}$ yr from Ref. [27].[6]

B. New limits for $0\nu2\beta$ decay of ^{116}Cd to ground state of ^{116}Sn

To estimate the half-life limit for the neutrinoless decay mode, a simple background model was used. In fact, in the $0\nu2\beta$ decay energy region only three background contribu-

[5]The radioactive impurities of all PMTs used in the installation were previously measured by R&D low background setup as (0.4–2.2) Bq/PMT and (0.1–0.2) Bq/PMT for ^{226}Ra and ^{228}Th activity, respectively [18].

[6]Note, that in Ref. [27] the quite small detection efficiency (1.73%) was calculated by the Monte Carlo method without experimental test, thus perhaps systematical error could be higher than quoted value.

TABLE I. Different origins of the systematical uncertainties and their contributions to the half-life value of ^{116}Cd two neutrino 2β decay.

Origin of the systematical error	Value range	Contribution to $T_{1/2}(2\nu)$ value, 10^{19} yr
Live measuring time	96^{+2}_{-8}%	+0.05, −0.2
Efficiency of PS analysis	98^{+1}_{-8}%	+0.05, −0.3
Detection efficiency of $2\nu2\beta$ decay (GEANT model uncertainty)	96±4%	±0.1
^{90}Sr–^{90}Y impurity in $^{116}CdWO_4$	≤0.17 mBq/kg	+0.5
^{234m}Pa impurity in $^{116}CdWO_4$	≤0.19 mBq/kg	+0.3

tions are important: (i) external γ background from U/Th contamination of the PMTs; (ii) tail of the $2\nu2\beta$ decay spectrum; and (iii) internal background distribution expected from the ^{212}Bi→^{212}Po→^{208}Pb decay (^{228}Th chain). As it was shown above, two decays in the fast chain ^{212}Bi→^{212}Po→^{208}Pb really create the background in the region of $0\nu2\beta$ decay [see Fig. 5(c)]. For the activity of ^{228}Th inside the $^{116}CdWO_4$ crystals two values were obtained: 38(3) μBq/kg (time-amplitude method) and 37(4) μBq/kg (pulse-shape analysis). Hence, in the limit of statistical errors, we do not find an indication of a failure for the rejection of α pulses by our PS analysis.

The high-energy part of the experimental spectrum of the $^{116}CdWO_4$ crystals measured in anticoincidence with the shielding detectors and after the time-amplitude and pulse-shape selection is shown in Fig. 8. The peak of $0\nu2\beta$ decay is absent, thus from the data we obtain a lower limit of the half-life: $\lim T_{1/2} = \ln 2 N\eta t/\lim S$, where $N = 4.66\times10^{23}$ is number of ^{116}Cd nuclei, t is the measuring time ($t = 4629$ h), η is the total detection efficiency for $0\nu2\beta$ decay, and lim$S$ is the number of events in the peak which can be excluded with a given confidence level. The value of the detection efficiency $\eta_{MC} = 0.83$ was calculated by the DECAY4 and GEANT3.21 codes, while the efficiency of the PS analysis $\eta_{PS} = 0.98$ was determined as described above, thus the total efficiency $\eta = \eta_{MC}\eta_{PS} = 0.81$. To obtain the value of lim$S$, the part of the spectrum in the 1.9–3.8 MeV region was fitted by the sum of the simulated $0\nu2\beta$ peak and three background functions (external γ rays from PMTs contamination; contribution from ^{212}Bi→^{212}Po→^{208}Pb intrinsic chain; and $2\nu2\beta$ decay tail). This fit gives the value of $S = -1.1\pm1.2$ counts, which corresponds—in accordance with Feldman-Cousins procedure for results close to the edge of physically accepted area [28] recommended by the Particle Data Group (PDG) [29]—to a lim$S = 1.0(0.4)$ counts with 90%(68%) C.L., and subsequently to $T_{1/2}(0\nu2\beta) \geq 1.3(3.7)\times10^{23}$ yr at 90%(68%) C.L. However, because of the low statistics in the energy range where the effect is expected, the obtained values can be cross checked in a more simple and explicit way. Indeed, in the energy interval 2.6–3.1 MeV (containing 91% of $0\nu2\beta$ peak) there is only one measured event, while the background expected on the basis of the GEANT simulation, in the same energy region is $3.2^{+2.1}_{-1.1}$ counts (1.9 ± 0.7 events from PMT contamination; 0.4 ± 0.1 events from $2\nu2\beta$ distribution; $0.9^{+2}_{-0.9}$ counts from mentioned ^{212}Bi→^{212}Po→^{208}Pb chain). Following the PDG recommendation [29,28] we can derive from these numbers the excluded limit as lim$S = 1.8(0.5)$ with 90%(68%) C.L., which leads to $T_{1/2}(0\nu2\beta) \geq 0.7(2.5)\times10^{23}$ yr at 90%(68%) C.L. confirming the previous estimate. Finally, the following values were accepted as conservative half-life limits for neutrinoless 2β decay of ^{116}Cd:

$$T_{1/2}(0\nu2\beta) \geq 0.7(2.5)\times10^{23}\ \text{yr},\quad 90\%(68\%)\ \text{C.L.}$$

Using calculations [17], one can obtain restrictions on the neutrino mass and right-handed admixtures in the weak interaction: $m_\nu \leq 3.0$ eV, $\eta \leq 3.9\times10^{-8}$, $\lambda \leq 3.4\times10^{-6}$ at 90% C.L., and neglecting right-handed contribution $m_\nu \leq 2.6(1.4)$ eV at 90% (68%) C.L. On the basis of calculations [27] we get a similar result: $m_\nu \leq 2.4(1.3)$ eV at 90%(68%) C.L. In accordance with Ref. [30] the value of the R-parity-violating parameter of minimal SUSY standard model is restricted by our $T_{1/2}$ limit to $\varepsilon \leq 8.8(6.4)\times10^{-4}$ at 90%(68%) C.L. [calculations [31] give more stringent restrictions: $\varepsilon \leq 3.4(2.4)\times10^{-4}$].

C. The bounds on $0\nu2\beta$ decay of ^{116}Cd to excited levels of ^{116}Sn

Not only ground state (g.s.) but also excited levels of ^{116}Sn with $E_{lev} \leq Q_{2\beta}$ can be populated in $0\nu2\beta$ decay of ^{116}Cd. In this case one or several γ quanta, conversion electrons and/or e^+e^- pairs will be emitted in a deexcitation process, in addition to two electrons emitted in 2β decay. The response functions of $^{116}CdWO_4$ detectors for $0\nu2\beta$ decay to the first and second excited levels of ^{116}Sn (2^+_1 with $E_{lev} = 1294$ keV and 0^+_1 with $E_{lev} = 1757$ keV) were simulated with the help of GEANT3.21 and DECAY4 codes. The full absorption of all emitted particles should result in the peak with $E = Q_{2\beta}$ (practically the same peak as it is expected for the $0\nu2\beta$ decay of ^{116}Cd to the g.s. of ^{116}Sn). Calculated full peak efficiencies are $\eta(2^+_1) = 0.14$ and $\eta(0^+_1) = 0.07$. These numbers and the value of lim$S = 1.8(0.5)$ with 90%(68%) C.L. (determined for the g.s.→g.s. transition) give the following restrictions on half-lives of ^{116}Cd neutrinoless 2β decay to excited levels of ^{116}Sn:

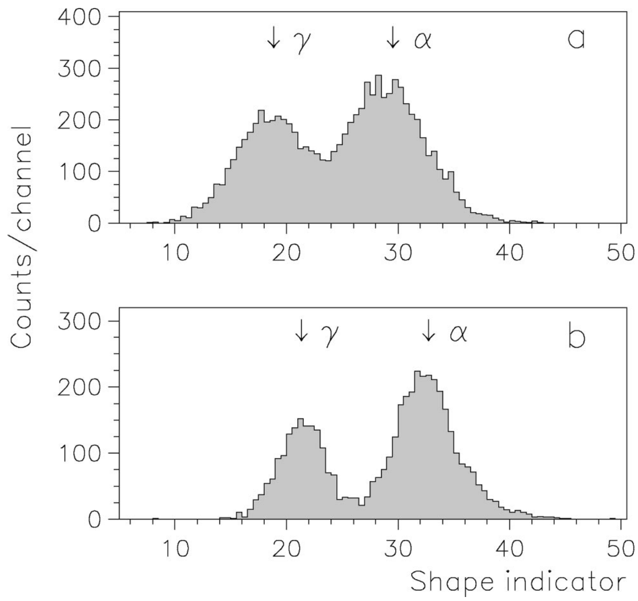

FIG. 9. Shape indicator (SI) distributions, which represent the PS discrimination ability of the $^{116}CdWO_4$ detectors: (a) for background events (0.8–1.2 MeV) collected during 4629 h before the last upgrading; (b) for background events (0.8–1.2 MeV) measured during 2734 h after the last upgrading.

$$T_{1/2}(\text{g.s.}\rightarrow 2_1^+)\geqslant 1.3(4.8)\times 10^{22}\ \text{yr},\quad 90\%(68\%)\ \text{C.L.},$$

$$T_{1/2}(\text{g.s.}\rightarrow 0_1^+)\geqslant 0.7(2.4)\times 10^{22}\ \text{yr},\quad 90\%(68\%)\ \text{C.L.}$$

D. Neutrinoless 2β decay with Majoron(s) emission

The procedure to obtain half-life limits for $0\nu2\beta$ decay with one (two) Majoron(s) emission was carried out in two steps as follows. First, because in the measured spectrum contributions of ^{40}K are negligible above the energy 1.6 MeV, the data were fitted in the energy region 1.6–2.8 MeV for $0\nu M1$ mode (1.6–2.6 MeV for $0\nu M2$) by using only three theoretical distributions: γ background from measured PMTs contamination (^{226}Ra and ^{232}Th chains) and two neutrino 2β decay of ^{116}Cd, as background, and $0\nu2\beta$ decay with one (two) Majoron(s) emission, as effect. With this simple model the χ^2 value was equal to 1.1 both for $0\nu M1$ and $0\nu M2$ fits. As a result, the number of events under a theoretical $0\nu M1$ curve was determined as 9 ± 21, giving no statistical evidence for the effect. It leads to an upper limit of 41(26) events at 90%(68%) C.L., that together with an efficiency value $\eta=0.905$ corresponds to the half-life limit $T_{1/2}(0\nu M1)\geqslant 3.7(5.9)\times 10^{21}$ yr. A similar procedure for $0\nu2\beta$ decay with two Majorons emission gives $T_{1/2}(0\nu M2)\geqslant 5.9(9.4)\times 10^{20}$ yr at 90%(68%) C.L. The part of the experimental spectrum and theoretical $0\nu M1$ and $0\nu M2$ distributions with half-lives equal to these limits are shown in Fig. 8. On one hand, the obtained results can be treated as conservative because the accepted background model consists of only two origins, the external background from U/Th contamination of the PMT and $2\nu2\beta$ decay distribution, while in the energy region of interest some other sources of background, such as the mentioned $^{212}Bi\rightarrow^{212}Po\rightarrow^{208}Pb$ chain contribution from the intrinsic impurities of the crystals, could enlarge the $0\nu M1$ and $0\nu M2$ limits. On the other hand, the uncertainty of the $2\nu2\beta$ decay half-life of ^{116}Cd could lead to the overestimated value of the $0\nu M1$ bound. To avoid the last possibility we have estimated the $T_{1/2}(0\nu M1)$ limit in a more explicit way. With this aim the energy interval 2.5–2.8 MeV—where the tail of $2\nu2\beta$ decay distribution is practically zero (see Fig. 8)—was used as the most sensitive region for the $0\nu M1$ double β decay search. In this energy interval the number of measured counts is 3, while the expected contribution (in the same energy range) from the PMT contamination is 3.2 ± 1.0 events and from $2\nu2\beta$ decay is 1.5 ± 0.5 events, thus the total expected background is 4.7 ± 1.1 counts. Following the PDG recommendation [29,28] we can derive from these numbers the excluded limit for the effect being sought as 3.0 events with 90% C.L. Taking into account that the interval 2.5–2.8 MeV contains 8.9% of full $0\nu M1$ curve, it yields a limit of $T_{1/2}(0\nu M1)\geqslant 4.5\times 10^{21}$ yr (90% C.L.), confirming the preceding estimate:

$$T_{1/2}(0\nu M1)\geqslant 3.7(5.9)\times 10^{21}\ \text{yr},\quad 90\%(68\%)\ \text{C.L.},$$

$$T_{1/2}(0\nu M2)\geqslant 5.9(9.4)\times 10^{20}\ \text{yr},\quad 90\%(68\%)\ \text{C.L.}$$

Both present half-life limits are more stringent than those established in our previous measurement during 19986 h [18] and in the NEMO experiment [27].

The probability of neutrinoless 2β decay with Majoron emission can be expressed as $\{T_{1/2}(0\nu M1)\}^{-1}=\langle g_M\rangle^2|\text{NME}|^2G$, where $\langle g_M\rangle$ is the effective Majoron-neutrino coupling constant, NME is the nuclear matrix element, and G is the kinematical factor. Using our result $T_{1/2}(0\nu M1)\geqslant 3.7(5.9)\times 10^{21}$ yr and values of G and NME calculated in the QRPA model with proton-neutron pairing [32] we obtain $g_M\leqslant 12(9.5)\times 10^{-5}$ [$g_M\leqslant 6.5(5.4)\times 10^{-5}$ on the basis of calculation [27]] with 90%(68%) C.L., which is one of the best restriction up-to-date obtained in the direct 2β decay experiments [1].

IV. CONCLUSION

The search for ^{116}Cd double β decay with enriched $^{116}CdWO_4$ scintillators has entered a new phase. The setup with four $^{116}CdWO_4$ crystals (339 g) has been running since October 1998. Improved passive shield, new active shield made of fifteen $CdWO_4$ crystals (total mass 20.6 kg), as well as time-amplitude and pulse-shape analysis of the data result in the reduction of the background rate in the 2.5–3.2 MeV region to 0.03 counts/yr kg keV. This reduction, together with an about threefold increase in the number of ^{116}Cd nuclei, leads to the substantial sensitivity enhancement of the ^{116}Cd experiment by more than one order of magnitude. Due to that the neutrino mass limit of $m_\nu\leqslant 2.6(1.4)$ eV at 90%(68%) C.L. was set after the first 4629 h run.

In August 1999 one of our $^{116}CdWO_4$ crystals was annealed at high temperature, and its light output was increased by $\approx$13%. The PMT of the main $^{116}CdWO_4$ detectors was changed by the special low background EMI tube (5 in. in diameter) with the RbCs photocathode, whose spectral response better fits the $CdWO_4$ scintillation light. As a result, the spectrometric parameters of four crystals taken as a whole were improved. In particular, the energy resolution of the main detector is now 11.4% at 1064 keV and 8.6% at 2615 keV (comparing with those before this upgrading: 14.5 and 11 %). In addition, the PS discrimination ability of the detector was improved too, as it is visible from Fig. 9, where the SI distribution of the measured background events—before and after the last upgrading—is depicted. It is expected that after approximately 5 yr of measurements the half-life limit $T_{1/2}(0\nu2\beta)\geqslant4\times10^{23}$ yr will be reached which corresponds to $m_\nu\leqslant1.2$ eV. The bounds on neutrinoless 2β decay with Majorons emission and 2β transitions to the excited levels of ^{116}Sn would be improved too.

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