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# Limits on Majoron modes of $^{116}\text{Cd}$ neutrinoless $2\beta$ decay

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## Abstract

Measurements of  $^{116}\text{Cd}$  double beta decay with the help of enriched  $^{116}\text{CdWO}_4$  crystal scintillators are in progress in the Solotvina Underground Laboratory. The last part of the exposition with the background rate 0.7 counts/y·kg·keV (in the region of  $^{116}\text{Cd}$   $0\nu 2\beta$  decay 2.7–2.9 MeV) is 22826 hours. For neutrinoless modes with emission of one and two Majorons, the limits  $T_{1/2}(0\nu M1) \geq 1.2 \times 10^{21}$  y and  $T_{1/2}(0\nu M2) \geq 2.6 \times 10^{20}$  y (90% C.L.) are determined. The above mentioned values give the restriction on the Majoron-neutrino coupling constant of  $g_M \leq 2.1 \times 10^{-4}$ . To improve these results, an advanced set-up with four enriched  $^{116}\text{CdWO}_4$  detectors shielded by fifteen large (1.5–2 kg)  $\text{CdWO}_4$  crystals has been mounted. © 1998 Elsevier Science B.V.

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## 1. Introduction

The exceptional interest in investigation of double beta ( $2\beta$ ) decay of atomic nuclei is explained by its fundamental importance [1–3]. Significant progress in the experimental techniques led to unambiguous direct observation of two-neutrino  $2\beta$  decay, a second-order process allowed by the Standard Model (SM) of electroweak interactions, for  $^{48}\text{Ca}$  [4],  $^{76}\text{Ge}$  [5],  $^{82}\text{Se}$  [6],  $^{100}\text{Mo}$  [7,8],  $^{116}\text{Cd}$  [9,10] and  $^{150}\text{Nd}$  [8,11], in addition

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to geochemical and radiochemical indications of  $2\beta$  processes in  $^{82}\text{Se}$  [12],  $^{96}\text{Zr}$  [13],  $^{128}\text{Te}$  [12,14],  $^{130}\text{Te}$  [12,14] and  $^{238}\text{U}$  [15] (see the compilation of Ref. [2] for further details and references). The values of  $T_{1/2}(2\nu 2\beta)$  measured in direct experiments lie in the region of  $10^{19}$ – $10^{21}$  y, and thus it is the rarest nuclear decay ever observed in the laboratory.

Neutrinoless  $2\beta$  decay is forbidden in the SM since it violates lepton number conservation. Nevertheless, many extensions of the SM predict a non-zero Majorana mass of the neutrino and, as a result,  $0\nu 2\beta$  processes. Despite considerable efforts devoted to the search for  $0\nu 2\beta$  decay in recent years, it still remains undetected. However, the impressive half-life limits in the range of  $10^{22}$ – $10^{23}$  y were established in direct experiments for  $^{82}\text{Se}$  [6],  $^{100}\text{Mo}$  [16],  $^{116}\text{Cd}$  [17],  $^{130}\text{Te}$  [18],  $^{136}\text{Xe}$  [19], and the highest limit was reached for  $^{76}\text{Ge}$ :  $T_{1/2}(0\nu 2\beta) \geq 1.1 \times 10^{25}$  y (90% C.L.) [20]. These results have surely shown the great potential of  $0\nu 2\beta$  processes to search for neutrino mass (as well as for right-handed admixture in the weak interaction, the neutrino-Majoron coupling constant, etc.) as a manifestation of new physics beyond the SM [21]. Moreover, as recently realized (see Refs. [21–23] and references therein), apart from the conventional light neutrino exchange mechanism,  $0\nu 2\beta$  decay can occur as a result of right-handed neutrino exchange in left–right symmetric models or via mechanisms based on the supersymmetric (SUSY) interactions (exchange of squarks, etc., in  $R$ -parity violating and exchange of sneutrinos in  $R$ -parity conserving SUSY models) and as a result of exchange of leptoquarks in models with leptoquarks. Therefore, at present  $0\nu 2\beta$  decay is considered to be a powerful test for different extensions of the SM, including some SUSY models, which could not only be complementary but in some cases competitive and even superior to the results of other running and forthcoming accelerator and non-accelerator experiments [21–23].

In some SM extensions double beta decay can occur also with the emission of a Majoron ( $M^0$ ), a hypothetical neutral pseudoscalar particle with zero mass, preferably interacting with Majorana neutrinos but not with ordinary substance. It was introduced in theory in 1980 [24] as a Goldstone boson associated with spontaneous breaking of lepton number (the Majoron of Ref. [24] was a singlet particle; a model with a triplet Majoron was proposed in Ref. [25] and with a doublet Majoron in Ref. [26]). This symmetry breaking gives rise to a non-zero neutrino mass. Interactions of  $M^0$  with charged leptons and quarks are strongly inhibited and appear only in higher orders of perturbation theory or as a result of  $M^0$  mixing with neutral Higgs bosons. Beside neutrino mass, the Majoron can also lead to other physical consequences, for example, to additional cooling of the Universe during the primordial nucleosynthesis or additional cooling of internal parts of stars.

Usually the triplet Majoron of Gelmini–Roncadelli [25] is regarded in connection with double beta decay. However, recent precise measurements of the  $Z^0$  boson width for decay into invisible particles showed that the number of light neutrino species is 3. The presence of a triplet Majoron would contribute two additional species to the  $Z^0$  width, thus a triplet Majoron (as well as a doublet) was ruled out. The incompatibility with the LEP data on the one hand and some experimental indications on the Majoron-like double

beta decays for several  $2\beta$  isotopes (which were diminished since that) on the other led to a theoretical re-examination and to a proposal of new Majoron models consistent with the LEP measurements (in particular, a massive vector Majoron, Majorons with lepton number  $L = -2$  or  $L = -1$ ) [27,28]. The schemes with emission of two Majorons in double beta decay were also proposed [29–31]. One can find nine different Majorons in the current theoretical literature (see the classification in Refs. [31,32]).

The Majoron modes of  $2\beta$  decay can be distinguished experimentally from the two-neutrino and neutrinoless  $2\beta$  decays by the different shapes of the energy spectra of the emitted electrons. The two-dimensional energy distribution is given by

$$\rho_{12}(e_1, e_2) = (e_1 + 1)p_1 F(e_1, Z)(e_2 + 1)p_2 F(e_2, Z)(Q_{\beta\beta} - e_1 - e_2)^n,$$

where  $e_i$  and  $p_i$  are the kinetic energy and momentum of the  $i$ th electron (in units of the electron mass  $m_0c^2$  and  $m_0c$ , respectively),  $F(e_i, Z)$  is the Fermi function, which takes into account the influence of the electric field of the nucleus on the emitted particles,  $Z$  is the atomic number of the daughter nucleus, and  $Q_{\beta\beta}$  is the energy release in  $2\beta$  decay. For double beta decay with emission of one Majoron the spectral index  $n$  is 1, for two Majorons  $n = 3$ , and for the usual two-neutrino  $2\beta$  decay  $n = 5$ . To obtain the energy spectrum for the sum of the electron energies,  $e = e_1 + e_2$ , the function  $\rho_{12}$  should be integrated numerically. However, in the Primakoff–Rosen approximation  $F(e_i, Z) \sim (e_i + 1)/p_i$ , which is quite good for  $2\beta^-$  decay, the integration can be performed in an analytical way, giving

$$\rho_{1+2}(e) = e(e^4 + 10e^3 + 40e^2 + 60e + 30)(Q_{\beta\beta} - e)^n,$$

with the same  $n$  as before. For  $0\nu 2\beta$  decay, the distribution of the sum of the electron energies is a  $\delta$ -function:  $\rho_{1+2}(e) = \delta(Q_{\beta\beta} - e)$ . (One of the modes with two-Majoron emission has spectral index  $n = 7$  [31], resulting in an even softer spectrum than that in  $2\nu 2\beta$  decay; it will not be regarded in the present work.)

The probability of  $2\beta$  decay with Majoron emission is

$$T_{1/2}^{-1} = \langle g_M \rangle^m |\text{NME}|^2 G,$$

where  $\langle g_M \rangle$  is the effective Majoron-neutrino coupling constant,  $m = 2$  for one- and  $m = 4$  for two-Majoron emission, NME is the nuclear matrix element, and  $G$  is a kinematical factor (different for different decay modes).

## 2. Experiment

With the aim to search for double beta processes in  $^{116}\text{Cd}$ , a cadmium tungstate crystal was grown with a mass of 510 g [33] enriched in  $^{116}\text{Cd}$  to 83%. The crystal was cleaved into five samples, three of which (19.0, 14.0 and 12.5 cm<sup>3</sup>; number of  $^{116}\text{Cd}$  nuclei is  $2.09 \times 10^{23}$ ,  $1.54 \times 10^{23}$  and  $1.37 \times 10^{23}$ , respectively) were used separately in different runs. All measurements, except the first one, were carried out in the Solotvina

Underground Laboratory (SUL) of the INR in a salt mine 430 m underground ( $\approx 1000$  m w.e., cosmic muon flux:  $1.5 \times 10^3 \text{ m}^{-2} \cdot \text{d}^{-1}$ , neutron flux:  $\leq 2.7 \times 10^{-6} \text{ cm}^{-2} \cdot \text{s}^{-1}$ , and radon concentration in air  $33 \text{ Bq} \cdot \text{m}^{-3}$ ) [34]. Various modifications of the installation were used in which the background rate in the vicinity of the energy release in  $2\beta$  decay of  $^{116}\text{Cd}$  ( $Q_{\beta\beta} = 2805(4) \text{ keV}$  [35]) was reduced successively by more than two orders of magnitude [17,33]. In the last modification, passive shielding of OFHC copper (5 cm), lead (23 cm) and polyethylene (16 cm) surrounds the plastic scintillator ( $\varnothing 38 \times 115 \text{ cm}$ ) of active shielding coupled to two low-background PMT FEU-125nf. The  $^{116}\text{CdWO}_4$  crystal is viewed by a PMT FEU-110 through a plastic scintillator light-guide 51 cm long. The energy resolution of the detector is 17.9, 10.1 and 8.0% at the energy of 662, 1770 and 2615 keV. The energy calibration was carried out with  $^{207}\text{Bi}$  weekly and  $^{232}\text{Th}$  once per fortnight. Residual gain shifts were corrected by software, and the resolution of background  $\gamma$  peaks did not differ significantly from that of the corresponding  $\gamma$  peaks in calibration runs (for the peak of 1461 keV ( $^{40}\text{K}$ ) FWHM= 11.6% for 19986 hours compared with 10.3% in calibration). The energy threshold was 30 keV for the  $^{116}\text{CdWO}_4$  detector and 200 keV for the active shielding and active plastic light-guide; the latter were measured before mounting of the set-up.

The event-by-event data acquisition system, based on an IBM PC compatible personal computer, allowed us to carry out measurements with up to 16 independent detectors simultaneously. For each event the following information was stored on hard disc: the amplitude (energy) of a signal, its arrival time and additional tags (coincidence–anticoincidence between different detectors, active shielding, cosmic  $\mu$  veto, etc.). The electronics and data acquisition system are described in more detail elsewhere [17,33].

The last improvement of the background was achieved by removing a thin (0.8–1.5 mm) external layer of the  $^{116}\text{CdWO}_4$  crystal of  $19.0 \text{ cm}^3$  (its volume decreased to  $16.2 \text{ cm}^3$  and, after repeated operation, to  $15.2 \text{ cm}^3$ ) where contamination by  $^{238}\text{U}$  was located, reducing it to  $\leq 20 \mu\text{Bq/kg}$  (50 times less than before). The background rate in the region of 2.7–2.9 MeV was 0.72 counts/y·kg·keV for 22826 h of data taking for  $^{116}\text{CdWO}_4$  crystals of 16.2, 15.2 and  $12.5 \text{ cm}^3$ . The experimental spectrum of  $^{116}\text{CdWO}_4$  crystal of  $15.2 \text{ cm}^3$  measured in anticoincidence with active shielding during 19986 h is shown in Fig. 1.

Now we pay attention to the trace radioactive contamination of the detector itself and the materials used in the set-up, because of the importance of these results for the reconstruction of the background components. The data were obtained by three different methods.

First, as shown previously [36], information on the arrival time of each event can be used for the analysis and selection of some decay chains in  $^{232}\text{Th}$ ,  $^{235}\text{U}$  and  $^{238}\text{U}$  families, like, for instance,  $^{214}\text{Bi}$  ( $Q_{\beta} = 3.3 \text{ MeV}$ )  $\rightarrow$   $^{214}\text{Po}$  ( $Q_{\alpha} = 7.8 \text{ MeV}$ ,  $T_{1/2} = 164.3 \mu\text{s}$ )  $\rightarrow$   $^{210}\text{Pb}$  or  $^{220}\text{Rn}$  ( $Q_{\alpha} = 6.4 \text{ MeV}$ )  $\rightarrow$   $^{216}\text{Po}$  ( $Q_{\alpha} = 6.9 \text{ MeV}$ ,  $T_{1/2} = 0.145 \text{ s}$ )  $\rightarrow$   $^{212}\text{Pb}$ . The energies of the first and second decays and the time interval between events are used to enhance the sensitivity and to reach better accuracy for detection of the trace radioactive contaminants in the detector. The events in the decay chain  $^{220}\text{Rn} \rightarrow ^{216}\text{Po} \rightarrow ^{212}\text{Pb}$ , selected in this way from the background, are shown in Fig. 2. The activities determined

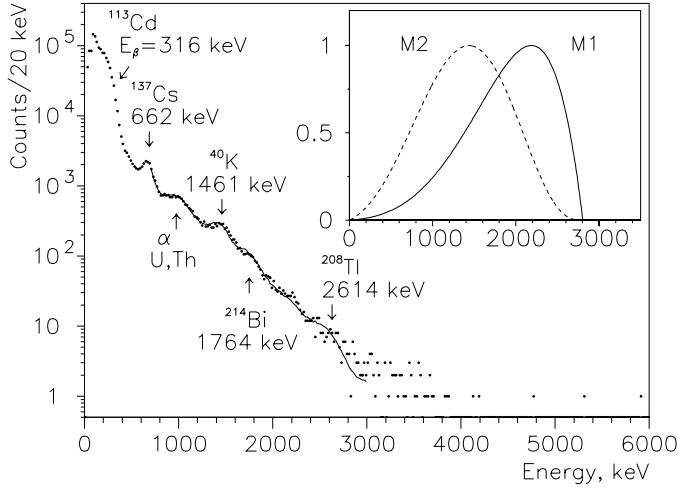


Fig. 1. Background spectrum of  $^{116}\text{CdWO}_4$   $15.2\text{ cm}^3$  measured during 19986 h in anticoincidence with active shielding (points). The calculated background in the energy region of the interest (700–3000 keV) is shown as a smooth curve (see Section 3 for details). In the insert, the theoretical distributions for the sum of the energies of the electrons in  $^{116}\text{Cd}$   $0\nu 2\beta$  decay with emission of one (M1) and two (M2) Majoron(s) are drawn.

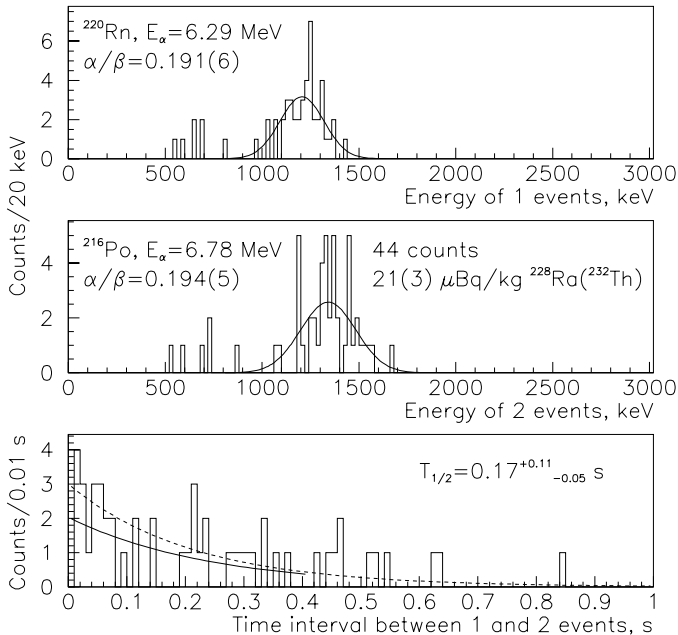


Fig. 2. (a,b) Energy spectra of first and second  $\alpha$  particles in the decay chain  $^{220}\text{Rn} \rightarrow ^{216}\text{Po} \rightarrow ^{212}\text{Pb}$  ( $^{116}\text{CdWO}_4$   $15.2\text{ cm}^3$ , 5130 h). Because of  $\alpha/\beta$  ratio  $\leq 1$ , the equivalent energy in the scale of  $\gamma$  quanta energies is approximately 5 times smaller. (c) Time distribution between the first and second events together with its fit by an exponent with  $T_{1/2} = 0.17\text{ s}$  (the table value  $T_{1/2} = 0.145\text{ s}$  [40] is shown as dashed line).

for the  $^{116}\text{CdWO}_4$  crystal of  $15.2\text{ cm}^3$  are as follows (in  $\mu\text{Bq/kg}$ ): 20(3) for  $^{228}\text{Ac}$  ( $^{232}\text{Th}$  family), 4(2) for  $^{227}\text{Ac}$  ( $^{235}\text{U}$  family),  $\leq 7.5$  for  $^{226}\text{Ra}$  ( $^{238}\text{U}$  family) and  $750_{-360}^{+220}$  for summary  $\alpha$  activity of U and Th families (see also Ref. [37]). Using this technique the relative light yield for  $\alpha$  and  $\beta$  particles  $\alpha/\beta = 0.148 + 0.0072E_\alpha$  ( $E_\alpha$  in MeV) and the energy resolution of the detector for  $\alpha$  particles  $\text{FWHM}_\alpha(E_\alpha) = 0.0444E_\alpha$  were also determined.

Secondly, the usual amplitude background spectrum, accumulated during 19986 h, was used to determine the activities (or their upper limits) of other internal radioactive contamination in the  $^{116}\text{CdWO}_4$  crystal as well as in the shielding (copper and plastic scintillator). To this aim, a simulation of the decay of various radioactive nuclides in the installation was performed with the help of GEANT 3.21 package [38]. The event generator [39] was used to describe the initial kinematics of the events (which particles and how many of them are emitted, what are their energies, directions of movement and times of emission). It takes into account the decay to the ground state as well as to excited levels of daughter nuclei with a subsequent complex de-excitation process [40]. The possibilities of emission of conversion electrons and  $e^+e^-$  pairs instead of  $\gamma$  quanta in nuclear transitions and angular correlation between emitted particles are taken into consideration. Simulated spectra of the  $^{116}\text{CdWO}_4$  crystal (in coincidence and in anticoincidence with active plastic shielding) were used to extract information on the presence of radioactive isotopes in the installation. For the  $^{116}\text{CdWO}_4$  crystal of  $15.2\text{ cm}^3$  itself, additional data on internal activities were determined as follows (in  $\text{mBq/kg}$ ): 3.2(2) for  $^{137}\text{Cs}$ ,  $\leq 2.0$  for  $^{40}\text{K}$  and  $\leq 0.8$  for  $^{90}\text{Sr}+^{90}\text{Y}$ . Data for copper and plastic scintillator are presented in Table 1.

Thirdly, some materials were measured with the R&D low-background set-up installed in the Solotvina Underground Laboratory where a natural  $\text{CdWO}_4$  detector (volume  $57\text{ cm}^3$ ) was used. Results of these measurements are also given in Table 1.

Table 1

Radioactive contamination of the materials used in the installation. All data (except for the plastic scintillator and OFHC copper) were obtained with the help of R&D set-up with natural  $\text{CdWO}_4$  detector  $57\text{ cm}^3$ . Activities are given in  $\text{Bq/kg}$ , apart from that for photomultipliers [ $\text{Bq/PMT}$ ] and mylar [ $\text{Bq/dm}^2$ ]

Material	$^{40}\text{K}$	$^{137}\text{Cs}$	$^{226}\text{Ra}$	$^{228}\text{Th}$
Plastic scintillator	$\leq 1.0 \times 10^{-2}$		$\leq 2.3 \times 10^{-3}$	$\leq 9.0 \times 10^{-5}$
OFHC copper	$\leq 2.8 \times 10^{-2}$		$\leq 3.7 \times 10^{-3}$	$\leq 2.5 \times 10^{-4}$
Lead	$\leq 4.0 \times 10^{-3}$	$\leq 1.4 \times 10^{-3}$	$\leq 2.0 \times 10^{-3}$	$\leq 1.0 \times 10^{-3}$
Boron polyethylene	$\leq 1.0 \times 10^{-2}$	$= 7.0(10) \times 10^{-2}$	$= 1.2(2) \times 10^{-2}$	$= 8.0(40) \times 10^{-4}$
Teflon	$\leq 4.0$	$\leq 0.5$	$\leq 1.5$	$\leq 1.2 \times 10^{-1}$
Optical grease (France)	$\leq 5.0 \times 10^{-1}$	$\leq 8.0 \times 10^{-2}$	$\leq 1.3 \times 10^{-1}$	$\leq 5.0 \times 10^{-2}$
Optical grease (USSR)	$\leq 1.0$	$\leq 9.0 \times 10^{-2}$	$\leq 6.0 \times 10^{-1}$	$\leq 6.0 \times 10^{-3}$
Mylar		$\leq 5.0 \times 10^{-5}$	$\leq 2.0 \times 10^{-5}$	$\leq 5.0 \times 10^{-6}$
PMT FEU-110	$= 3.0(3)$		$= 0.8(2)$	$= 0.17(7)$
PMT FEU-125nf	$= 3.0(2)$		$= 2.2(9)$	$= 0.20(3)$

### 3. Results and discussion

The limits on  $2\beta$  decay modes with emission of one Majoron ( $0\nu M1$ ; spectral index in the formulae for energy distributions  $n = 1$ ) or two Majorons ( $0\nu M2$ ; spectral index  $n = 3$ ) can be obtained in the simplest and very conservative way by just demanding that, in any energy region, theoretical Majoron(s) distributions can not exceed the experimental spectrum. There are 60 events in the region of 2500–2660 keV. Attributing all these events to  $0\nu M1$   $2\beta$  decay and using the relation  $T_{1/2} = N \cdot \varepsilon \cdot t \cdot \ln 2 / S$ , where  $N$  is number of  $^{116}\text{Cd}$  nuclei ( $N = 1.67 \times 10^{23}$ ),  $t$  is the time of the measurements ( $t = 19986$  h),  $\varepsilon$  is efficiency ( $\varepsilon = 1$ ) and  $S$  is number of events under full Majoron curve ( $S = 745$ ), we obtain  $T_{1/2}(0\nu M1) = 3.5 \times 10^{20}$  y. However, the real  $T_{1/2}$  should be higher because part of the events is certainly caused by decay of the radioactive impurities in various components of the installation. To demonstrate this statement, we can take into account some contributions from impurities for which values (not upper limits) were measured, in particular

- (1)  $^{228}\text{Ac}$  chain in the  $^{116}\text{CdWO}_4$  crystal (determined by a time–amplitude analysis of events): activity of  $20 \mu\text{Bq/kg}$  gives 10.5 counts in the region of 2500–2660 keV;
- (2) impurities in the photomultipliers FEU-110 and FEU-125nf (measured in the set-up with the natural  $\text{CdWO}_4$  detector  $57 \text{ cm}^3$ ). Total activity of 5.2 Bq for the  $^{226}\text{Ra}$  chain in two PMTs FEU-125nf and one PMT FEU-110 gives 4.3 counts, and a total activity of 0.57 Bq for the  $^{228}\text{Th}$  chain gives 13.8 counts in the same energy region;
- (3) two-neutrino double beta decay of  $^{116}\text{Cd}$  has been recently observed by the Osaka–Kiev collaboration with  $T_{1/2}(2\nu 2\beta) = (2.6_{-0.6}^{+0.9}) \times 10^{19}$  y [9] and by the NEMO collaboration with  $T_{1/2}(2\nu 2\beta) = (3.8 \pm 0.4) \times 10^{19}$  y [10]. If we accept the latter value, we obtain 0.8 counts in the 2500–2660 keV region.

Subtracting only these three contributions from our 60 counts in the experimental spectrum (see Fig. 3), we obtain 30.6 counts which corresponds to  $T_{1/2}(0\nu M1) = 6.9 \times 10^{20}$  y.

For double beta decay with two Majorons emission, in the energy region of 1880–2100 keV there are 460 events in the experimental spectrum. All attributed to  $0\nu M2$   $2\beta$  decay, they give  $T_{1/2}(0\nu M2) = 5.7 \times 10^{19}$  y. The contribution from the three above-mentioned sources are: (1) from  $^{232}\text{Th}$  chain in crystal 10.2 counts; (2) from  $^{228}\text{Th}$  chain in three PMTs 5.1 counts, and from  $^{226}\text{Ra}$  chain in three PMTs 36.9 counts; (3) from  $2\nu 2\beta$  decay of  $^{116}\text{Cd}$  (if  $T_{1/2}(2\nu 2\beta) = 3.8 \times 10^{19}$  y) 205.7 counts. Subtracted from 460 original events, these contributions result in  $T_{1/2}(0\nu M2) = 1.3 \times 10^{20}$  y (Fig. 3).

The obtained values of  $T_{1/2}(0\nu M1)$  and  $T_{1/2}(0\nu M2)$  are still conservative because there are other background contributions in the corresponding energy regions. For example, if we suppose the same activity of radon in the remnant cavities inside our installation as in the air inside the laboratory ( $33 \text{ Bq/m}^3$ ), we could receive 36 counts in the 2500–2660 keV region, even more than our remaining number of events (30.6) there. However, because the activity inside the installation is surely lower than in free air in the laboratory, the amplitude of the radon response function was treated as a free

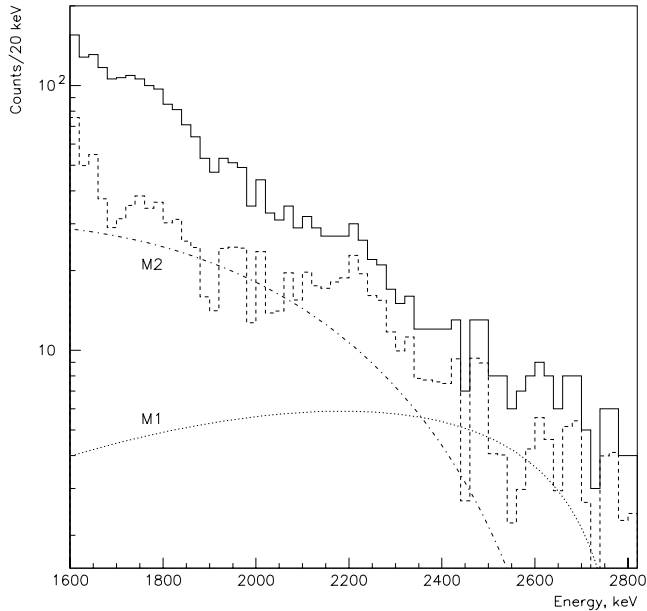


Fig. 3. Solid histogram: the experimental spectrum of  $^{116}\text{CdWO}_4$   $15.2 \text{ cm}^3$ , 19986 h in the energy region of 1600–2800 keV. Dashed histogram: the experimental spectrum with subtracted contributions from PMTs,  $^{228}\text{Th}$  chain in the  $^{116}\text{CdWO}_4$  crystal and  $2\nu2\beta$  decay of  $^{116}\text{Cd}$ . The smooth curve M1 (M2) is the theoretical curve for  $^{116}\text{Cd}$   $0\nu2\beta$  decay with one-(two-)Majoron emission with  $T_{1/2} = 6.9 \times 10^{20}$  ( $1.3 \times 10^{20}$ ) y.

parameter (restricted by the value of  $33 \text{ Bq/m}^3$  + two error bars) afterwards during the fitting procedure in which the amplitude of the Majoron spectrum was rectified.

To take into account the contributions from other sources, for activities of which the upper limits were only established, we used the simulation of various radioactive contamination in the installation. The response functions (RFs) of the  $^{116}\text{CdWO}_4$  detector (in anticoincidence and coincidence with active shielding) were calculated with GEANT 3.21 [38] (and event generator [39]) for possible internal sources ( $^{113}\text{Cd}$ ,  $^{113m}\text{Cd}$ ,  $^{40}\text{K}$ ,  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}+^{90}\text{Y}$ , chains of  $^{232}\text{Th}$  and  $^{238}\text{U}$ ) as well as for external sources of  $^{40}\text{K}$ ,  $^{232}\text{Th}$  and  $^{238}\text{U}$  chains located in photomultipliers, shielding of plastic, Cu and Pb, materials in close vicinity of  $^{116}\text{CdWO}_4$  crystal (mylar, teflon and optical grease),  $^{220}\text{Rn}$  in air in the shielding cavities and bremsstrahlung from  $\beta$  decay of  $^{210}\text{Bi}$  (daughter of  $^{210}\text{Pb}$ ) in Pb. The original (without any subtraction) coincidence (measured during 11420 h) and anticoincidence (19986 h) experimental spectra were fitted simultaneously by a set of simulated RFs for radioactive sources and  $^{116}\text{Cd}$   $2\nu2\beta$  decay and RF for being sought  $0\nu\text{M1}$  (or  $0\nu\text{M2}$ )  $2\beta$  decay. Some RF amplitudes were fixed (restricted) for known content (upper limit) of radioactive contamination in the material (see previous section and Table 1 for the values). To take into account possible errors (due to ambiguities in source location, etc.), we increased the error bars of the determined RF amplitudes by a factor of 2. Data on the shape of the fourth forbidden  $\beta$  decay of  $^{113}\text{Cd}$ , which is present in the enriched  $^{116}\text{CdWO}_4$  crystal on the level of 2.15% [33], were taken from



Ref. [41]. The amplitude of  $^{116}\text{Cd}$   $2\nu 2\beta$  decay in our fit was restricted between the values of  $2.0 \times 10^{19}$  y and  $4.2 \times 10^{19}$  y (the values and error bars of Refs. [9,10] were taken into account).

From the fit (for which we used the PAW package [42]) in the energy region of 700–3000 keV, the number of events under the theoretical curves for  $0\nu M1$  and  $0\nu M2$   $2\beta$  decays was determined as  $84 \pm 96$  and  $510 \pm 385$  respectively, giving no statistical evidence for the effect. The value of  $\chi^2$  was equal to 1.0 for  $0\nu M1$  and 0.91 for  $0\nu M2$ . Using these values, the upper limits on the events number  $S$  were calculated in accordance with the Particle Data Group procedure for renormalization of the probability function in a physically acceptable area [43]:  $S(0\nu M1) \leq 218$  and  $S(0\nu M2) \leq 1024$  (90% C.L.). The calculated limits on the half-life are:  $T_{1/2}(0\nu M1) \geq 1.2 \times 10^{21}$  y and  $T_{1/2}(0\nu M2) \geq 2.6 \times 10^{20}$  y (90% C.L.). As one can see, the fitting procedure allowed us to improve the half-life limit 1.7 times for  $0\nu M1$  and 2.0 times for  $0\nu M2$   $2\beta$  decay. The first value is the same as that obtained by the NEMO Collaboration [10], the second result is determined for the first time.

For neutrinoless  $2\beta$  decay with two-Majoron emission, our limit  $T_{1/2}(0\nu M2) \geq 2.6 \times 10^{20}$  y (90% C.L.) can be compared only with two other limits known to date: for  $^{100}\text{Mo}$   $T_{1/2}(0\nu M2) \geq 5.3 \times 10^{19}$  y with 68% C.L. [44] and for  $^{76}\text{Ge}$   $T_{1/2}(0\nu M2) \geq 5.9 \times 10^{21}$  y with 90% C.L. [45]. The obtained value can be used to derive the constraint on the mass of the Zino, the superpartner of the  $Z^0$  boson in supersymmetric models with spontaneous violation of  $R$ -parity, using the relation [29]

$$T_{1/2}^{-1} = \langle g_{MM} \rangle^2 |\text{NME}|^2 G,$$

where the coupling constant  $\langle g_{MM} \rangle$  is connected with the mass of the Zino  $M_Z$  and the Weinberg angle  $\theta_W$ :  $g_{MM} = g^2/4M_Z \cos^2 \theta_W$  and  $g = \sqrt{4\pi\alpha}/\sin \theta_W = 0.406$  is the SU(2) gauge coupling constant. However, as in Ref. [44], the calculated limit  $M_Z \gtrsim 0.05$  GeV is not competitive with that obtained in accelerator experiments:  $M_Z \gtrsim 30$  GeV [43].

Experimental limits on  $T_{1/2}(0\nu M1)$  for various  $2\beta$  isotopes established in direct measurements lay in the range of  $10^{17}$ – $10^{22}$  y [1,2]. Using our result  $T_{1/2}(0\nu M1) \geq 1.2 \times 10^{21}$  y and the relation between  $T_{1/2}$  and  $\langle g_M \rangle$  given in Section 1, we can deduce an upper limit for the effective Majoron-neutrino coupling strength  $\langle g_M \rangle$ . With the phase space integral  $G$  and the nuclear matrix element NME calculated in the QRPA model with proton–neutron pairing [32] we obtain  $g_M \leq 2.1 \times 10^{-4}$ . Using the  $G$  and NME computed in the QRPA scheme of Ref. [46], one deduces  $g_M \leq 3.1 \times 10^{-4}$  (identical results with and without p–n pairing). Finally, if we use the same  $G$  and NME of Ref. [47] as in Ref. [10], we obtain the slightly better value of  $g_M \leq 1.2 \times 10^{-4}$ . Thus we can conclude that the upper limit on  $\langle g_M \rangle$  lies in the range of  $(1 - 3) \times 10^{-4}$ , as for most of the other  $2\beta$  nuclides investigated in direct experiments [1].

It is interesting to compare the limits derived from  $2\beta$  decay experiments with the constraints on  $g_M$  obtained by other methods.

The presence of Majorons during big-bang nucleosynthesis and their additional degrees of freedom would lead to a faster expansion and cooling of the universe and an earlier freezing of non-decayed neutrons in deuterium and  $^4\text{He}$  nuclei, therefore resulting

in a larger  ${}^4\text{He}$  abundance. The observed  ${}^4\text{He}$  abundance thus gives  $g_M \leq 9 \times 10^{-6}$  [48]. Majoron-like particles can also influence the dynamics of supernova explosions, and from the data on the SN1987 neutrino flux the limit  $g_M \leq 3 \times 10^{-4}$  was deduced in Ref. [49]. Note however that these values are not model independent and can not be compared with  $2\beta$  data directly.

On the other hand, restrictions obtained in direct measurements of semileptonic decays of  $\pi$  and  $K$  mesons  $\pi(K) \rightarrow l + \nu + M^0$  ( $l$  is a lepton) are much less severe ( $g_M \lesssim 10^{-2}$  [43]) compared to those from  $2\beta$  decay research.

Considering these circumstances we plan to improve our experiment on  ${}^{116}\text{Cd}$   $2\beta$  decay further. In order to enhance the sensitivity, the following improvements of the installation are scheduled: increase of the number of  ${}^{116}\text{Cd}$  nuclei, reduction of the background and improvement of data taking and processing. Three additional  ${}^{116}\text{CdWO}_4$  crystals have been mounted now in order to increase the number of  ${}^{116}\text{Cd}$  nuclei. An additional active shielding made of fifteen natural  $\text{CdWO}_4$  crystals of large volume ( $\simeq 200 \text{ cm}^3$  each) [37] is installed in the very neighborhood of the main enriched detectors screening them fully in order to reduce the background rate. Due to the high purity of the natural  $\text{CdWO}_4$  crystals [37] and their density of  $\simeq 8 \text{ g/cm}^3$ , the  $\text{CdWO}_4$  shielding of 6–7 cm thickness will result in a further suppression (up to 5 times) of the  ${}^{116}\text{CdWO}_4$  background in the energy region of interest.

An additional possibility, which can also reduce the background, is the pulse shape discrimination (PSD) technique with  $\text{CdWO}_4$  crystal scintillators. To this aim, in collaboration with INFN (Florence), the properties of light emission from  $\text{CdWO}_4$  were investigated with different kinds of sources. A clear discrimination between  $\gamma$  rays and  $\alpha$  particles was achieved [50], which allows us to use this technique in the measurements. For applying this method in the experiment, the new complementary data acquisition system was developed on the basis of flash-ADC (Analog Devices AD9022), parallel digital I/O board (PC-DIO-24) and IBM PC [50] which are mounted in the Solotvina Underground Laboratory now.

#### 4. Conclusions

In this work the half-life limits on the  $0\nu 2\beta$  decay of  ${}^{116}\text{Cd}$  with one- and two-Majoron emission are presented:  $T_{1/2}(0\nu M1) \geq 1.2 \times 10^{21} \text{ y}$  and  $T_{1/2}(0\nu M2) \geq 2.6 \times 10^{20} \text{ y}$  (90% C.L.). The last one is determined for the first time. The corresponding constraints on the Majoron-neutrino coupling constant are in the range of  $g_M \leq (1-3) \times 10^{-4}$ , better than those obtained in direct measurements of semileptonic decays of  $\pi$  and  $K$  mesons ( $g_M \lesssim 10^{-2}$ ).

This fact strongly supports the further enhancement of the sensitivity in the current  $2\beta$  decay experiment with  ${}^{116}\text{Cd}$ , which is being performed now. It includes the increase of the number of  ${}^{116}\text{Cd}$  nuclei by a factor of 3 (three additional  ${}^{116}\text{CdWO}_4$  crystals), reduction of the background by a factor of up to 5–8 (new active shielding made of fifteen large  ${}^{nat}\text{CdWO}_4$  crystals) and improvement in data acquisition and processing

(time analysis, pulse shape discrimination, etc.). We expect that all these measures will provide the possibility to reach the limit of  $T_{1/2}(0\nu\text{M1}) \geq (0.6 - 1.0) \times 10^{22}$  y, which corresponds to  $g_M \leq (0.4 - 1.0) \times 10^{-4}$ .

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