#### Double Beta Decay Experiments at Kiev

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ABSTRACT: Search for double beta decay of <sup>116</sup>Cd and <sup>196</sup>Hg was made in the Solotvina underground laboratory operating at 1000 m w.e.

#### 1. INTRODUCTION

The underground laboratory of the INR (Kiev) was built in the operating Solotvina salt mine. The Laboratory is situated 430 m deep from the earth surface that corresponds to 1000 m w.e. Cosmic ray flux is reduced by a factor of about  $10^4$ . Due to a low radioactive contamination of salt the natural  $\gamma$ -background in Solotvina Laboratory is 10-100 times lower than in other underground laboratories [1]. Low background scintillation and semiconductor detectors were used successfully for investigations of double beta decay and other rare processes.

## 2. SEARCH FOR 28 DECAY OF 116cd

The energy released in the  $2\beta$  transition  $^{116}\text{Cd}-^{116}\text{Sn}$  is  $^{2802}(4)$  keV [2]. To search for  $2\beta$  decay a  $^{Cd}\text{WO}_4$  scintillator enriched by  $^{116}\text{Cd}$  to 83% has been used [3]. Samples of metallic Cd were obtained from the State Foundation (619 g, 91%). After purification by vacuum distillation Cd was put in the form of the oxide CdO. A cadmium wolframite single crystal with a mass of 510 g (11.2 cm long, max diameter 3.5 cm) was grown by the Czochralski technique from a composite material prepared by sintering a mixture of the oxides WO4 and CdO. The crystal was then cleaved into five samples and the best one (3.4 cm in diameter, 2.2 cm height) was used in the measurements. The number of  $^{116}\text{Cd}$  nuclei in this sample is  $^{2.18}\text{x}_{10}^{23}$ .

The first experiment was carried out with a simple counter consisting of a photomultiplier, a quartz light pipe (5 cm diameter, 5 cm long) and the crystal. The energy resolution of the detector was 16% and 9% at the energy of 662 and 2615 keV, respectively. The detector was surrounded by a shield of mercury (8 cm thick), lead (23 cm) and polyethylene (32 cm). After 1016.8 h of counting at the Solotvina underground

laboratory the lower limit of  $1.3 \times 10^{21}$  y was found for the neutrinoless  $2\beta$  decay half-life of  $^{116}\text{Cd}$  (at the confidence level of 68%) [3].

The set up was then changed in order to improve the sensitivity. Two crystals (the enriched -  $19.2~\rm cm^3$  and the natural -  $56.5~\rm cm^3$ ) were coupled to photomultipliers through the light pipe 9 cm long. The active shielding of the detectors consists of four well-type CsI crystals with diameter of 15 cm and the full length of 40 cm. The  $^{116}{\rm CdWO_4}$ ,  $^{16}{\rm CdWO_4}$  and CsI scintillators were shielded by high purity mercury (8 cm thick), lead (23 cm) and polyethylene (24 cm). The plastic scintillator (110x110x6 cm<sup>3</sup>) was placed under set up and operated as an antimuon shielding.

The measured spectra are shown in Figures 1 and 2 (the enriched crystal - 2588 h, the natural one - 2525 h). The peaks observed in the spectra are caused mainly by  $^{40}$ K and  $^{208}$ Tl from PMT's and  $^{137}$ Cs from CsI counters. There are two differences in our spectra: an increase in counts below 320 keV for the natural crystal and a small peak at the energy of about 800 keV for the enriched crystal. The first difference can be attributed to the fourth-forbidden  $^{6}$  decay of  $^{113}$ Cd with half-life of about  $^{106}$  y [2]. The second one can be caused by  $^{67}$ particles from an intrinsic radioactive contamination of the enriched scintillator. We measured the  $^{67}$ background of the crystals and found the spectrum of the enriched crystal likes the  $^{67}$ spectrum of  $^{238}$ U at the contamination level of about  $^{11}$ Cl  $^{10}$  g/g. Only limits were set for the presence of  $^{232}$ Th  $^{23}$ Cl  $^{23}$ Cl and  $^{226}$ Ra  $^{23}$ Cl  $^{23}$ Cl

Within the energy region of 0 116Cd (2676-2928 keV) the background rate of 116CdWO $_4$  is  $7\times10^{-6}$  counts/h/keV/cm $^3$ . Using this rate, number of nuclei and time of measurement, the limits were calculated for half-life of  $2\beta$  decay of 116Cd:

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0 \mathbf{r}2\beta \sim (3-5) \times 10^{21} y (68\% \text{ C.L.}) 0 \mathbf{r}2\beta, M \sim 1.0 \times 10^{21} y (68\% \text{ C.L.}) 2 \mathbf{r}2\beta \sim 4.5 \times 10^{18} y (99\% \text{ C.L.})
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Comparing our experimental result with theory [4] we find the limit on neutrino mass to be less than 8 eV. Now we plane to use four  $^{11\odot}\text{CdWO}_4$  crystals, to improve the energy resolution and to reduce the background of the detectors. We hope to reach the half-life limits of  $10^{23}$  years and the corresponding neutrino mass of about 1.5 eV.

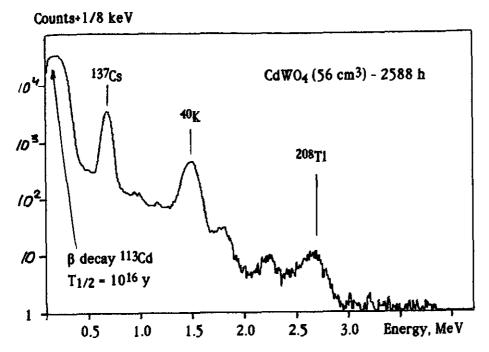


Fig. 1.  $CdWO_4$  background spectrum for 2588 h

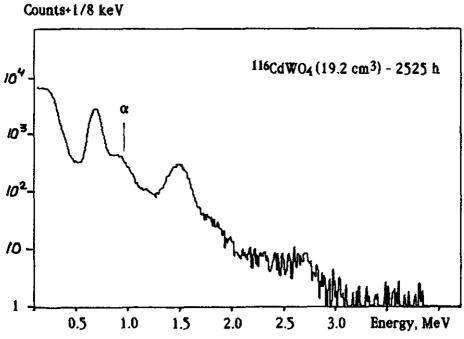


Fig. 2. 116CdWO $_4$  background spectrum for 2525 h

## 3. LOW BACKGROUND HPGe DETECTOR AND RARE DECAYS OF MERCURY

This work was made in collaboration with Max Plank Institut fuer Kernphysik (Heildelberg, Germany) [5,6]. The intrinsic p-type Ge coaxial detector (PGT) (the active volume of 165 cm $^3$ , the relative efficiency of 38.7% and the energy resolution FWHM of 1.9 keV at the 1333 keV) was used. The Ge crystal without any construction units was overpacked into a low background cryostat, which was made of titanium and oxygen free copper. The cooled stage of the pre-amplifier was situated into the cryostat. Finally the detector had following characteristics: the efficiency - 35(4)%, FWHM - 2.7 keV, the operating bias - 2000 V.

The first layer of the passive shielding is mercury, which encloses the detector completely. Inside the cryostat a container with mercury is placed too. A mercury thickness is changing from 9 to 30 cm. Then the layers of oxygen free copper (11-15 cm), lead (23-30 cm) and polyethylene (24 cm) follows. The full weight of Pb is equal to 12000 kg, Cu - 1800 kg, Hg - 570 kg. The overall shield thickness is equal to 620 g/cm² from the upper part, 570 g/cm² at sides and 600-650 g/cm² from bottom. The veto antimuon shielding consists of plastic scintillator of 117x114x9 cm³. The protection against electromagnetic induction and 'microphonic' noise was in action too [5].

As a result the background rate of the detector within 100-2850 keV was equal to 20.6 counts/h and within the energy region of 0\*28 decay of 76Ge was equal to 2.4 counts/y/keV/kg that corresponds to the best low background HPGe detectors. All peaks in the spectrum have been identified: they belong  $_{\text{to}}$  40K. 134,137Cs and to 235,238U and 232Th family members. For instance the intensity of the 2615 keV peak from  $^{208}{\rm Tl}$  was about 0.06 counts/h. A breaking of secular equilibrium in the 238U family has been observed: peaks of 226Ra and its daughter products were absent. This may take place in metals as a result of technological processes [7,8]. We calculated the impurities of Ti (walls of the cryostat and containers):  $2x10^{-9}$  g/g - 232Th;  $2x10^{-10}$  g/g - 235U;  $1.6x10^{-8}$  g/g - 238U (only  $234_{\mathrm{Th}}$ ). These levels are in good accordance with previous measurements [8]. Now we have made the cryostat from silicon of the semiconductor purity and hope to reach the background rate less than 1 count/y/keV/kg in the energy region of 0728 decay of  $^{76}\mbox{Ge}.$  But even the existing background and 570 kg of mercury placed around the detector allows to establish limits on the probability of some rare decays of mercury.

 $^{196}\mbox{Hg}$  can undergo, e.g. a double electron capture. The abundance of  $^{196}\mbox{Hg}$  is 0.15%, atomic mass difference  $^{196}\mbox{Hg}$  –  $^{196}\mbox{Pt}$  is equal to 820(3) keV. Taking into account the K-

electron binding energy and the OM2K capture energy (663.2 keV) the nucleus may be de-exciting by two Y-quanta with energy of 307.5 and 355.7 keV. These peaks are absent in the background spectrum for 1108.6 h (Figure 3) and the average rate in the region of 300-400 keV is equal to 1.8(0.1)x10<sup>-2</sup> counts/h/keV. The data in the vicinity of possible peaks are fitted by the least-square method as sum of two functions: a linear (background) and Gaussian (effect) with given position and variance. It was obtained that the peak area at the energy of 307.5 keV doesn't exceed 13 counts and at 355.7 keV - 6 counts (C.L. 68%). The calculated efficiency is equal to 6.5x10<sup>-4</sup> (307.5 keV) and 7.7x10<sup>-4</sup> (355.7 keV). It leads to the following estimates of 196Hg half-life:

Lim  $T_{1/2}$  (0)2K) =9.6x10<sup>17</sup> years (E<sub>y</sub>=307.5 keV)

Lim  $T_{1/2}$  (0#+2#)=2.5x10<sup>18</sup> years (E $_{\gamma}$ =355.7 keV) These values 7 and 16 times exceed the limits determined earlier [9].

# Counts/0.5 keV 212ph 238.6 HP Ge 165 cm3 - 1108.6 h 100 137Cs 80 661.7 60 307.5 355.7 208TI 510.6 583.2 0 300 600 Energy, keV 200 400 500

Fig. 3. HPGe detector background spectrum

It is known that the pion condensate in nuclear matter can result in a second minimum for the dependence of nucleus energy on its density. If this minimum corresponds to a density and binding energy higher that in an ordinary nucleus the nucleus may transit in superdense state

spontaneously [10]. Lifetimes limits  $(3x10^{21}-10^{24} \text{ y})$  were measured [11] for some nuclei except Hg. In our work [6] we attempted to detect a high energy y-radiation from 570 kg  $(1.72x10^{27} \text{ nuclei})$  of mercury which may accompany the hypothetical transitions [11].

The background spectrum in the range of 0.5-26 MeV measured during 805.9 h is presented in Figure 4. The background rate above 4 MeV is extremely low:  $7.5 \times 10^{-6}$ ;  $1.5 \times 10^{-5}$ ;  $(1-3) \times 10^{-6}$  counts/h/keV at the energy region 4-5; 5-5.5 and 6-26 MeV, respectively. Taking into account the efficiency, the following limit of Hg lifetime concerning a transition into a superdense state is obtained:

Lim  $\tau = (0.8-1.5) \times 10^{22}$  years (95% C.L.)

### Counts+1/50 keV

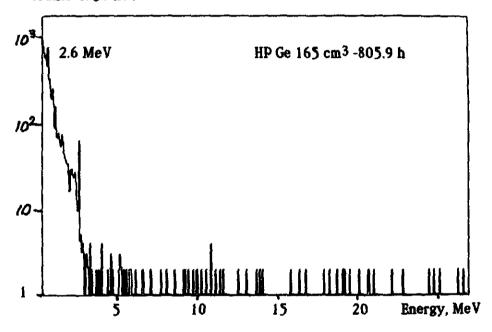


Fig. 4. HPGe background spectrum in the interval 0.5-26~MeV

The discovery of cluster radioactivity [12] in 1984 excited a great interest to this phenomenon. Several nuclides decaying with  $^{14}\mathrm{C}$ ,  $^{24}\mathrm{Ne}$  and  $^{28}\mathrm{Mg}$  emission have been found. For a better understanding of this process it is desirable to enlarge the number of investigated nuclei. With this purpose limits on probability of mercury decay with  $^{24}\mathrm{Ne}$  and  $^{28}\mathrm{Mg}$  emission have been studied in our laboratory [6].

The cascade emission of two  $\gamma$ -quanta (2754.0 keV and 1368.6 keV) is a final result of  $^{24}{\rm Ne}$  decay. The  $^{28}{\rm Mg}$  decay is

accompanied by Y-quantum of 1779.0 keV. Peaks with these energies have not been detected. The half-life limits for various mercury isotopes calculated with the confidence level of 95% are lying between (1.3-3.7)x10<sup>21</sup> y. It should be noted that these are the highest limits obtained in experimental searches of cluster radioactivity.

#### REFERENCES

- [1] Zdesenko Yu G et al 1988 Proc. Int. Symp. Underground Physics (Baksan Valley, 1987) ed G V Domogatsky (Moscow, Nauka) 291
- [2] Lederer C M and Shierly V S 1978 Table of Isotopes (New York, Wiley)
- [3] Danevich F A et al 1989 Pis'ma JETP 49(8) 417
- [4] Grotz K and Klapdor H V 1986 Il Nuovo Cimento 9 C 535
- [5] Vishnevsky I N et al 1990 Proc. USSR Symp. on 26 decay problem (Kiev, 1989) ed Yu Zdesenko (Kiev, INR)
- [6] Buchner E et al 1990 Yad. Fiz. 52(2) 305
- [7] Liquory C et al 1983 Nucl. Instr. Meth. 204 585
- [8] Gavrin V N et al 1986 Preprint INR (Moscow) P-0494
- [9] Zdesenko Yu G and Kuts V N 1986 Pis'ma JETP 43(10) 459 [10] Mishustin I N and Karnyuhin A V 1980 Yad. Fiz. 32(10)
- [11] Aleshin V I et al 1976 Pis'ma JETP 24(2) 114 Aleshin V I et al 1979 Preprint IAE (Moscow) 3127
- [12] Rose H J and Jones G A 1984 Nature 307 24 Alexandrov D V et al 1984 Pis'ma JETP 40 152