

# Search for double beta decay of zinc and tungsten with low background ZnWO<sub>4</sub> crystal scintillators

R. Bernabei<sup>1,2</sup>, P. Belli<sup>2</sup>, F. Cappella<sup>3,4</sup>, R. Cerulli<sup>5</sup>, F.A. Danevich<sup>6</sup>, B.V. Grinyov<sup>7</sup>, A. Incicchitti<sup>4</sup>, V.V. Kobychiev<sup>6</sup>, V.M. Mokina<sup>6</sup>, S.S. Nagorny<sup>6</sup>, L.L. Nagornaya<sup>7</sup>, S. Nisi<sup>5</sup>, F. Nozzoli<sup>1,2</sup>, D.V. Poda<sup>6</sup>, D. Prospero<sup>3,4</sup>, V.I. Tretyak<sup>6</sup>, S.S. Yurchenko<sup>6</sup>

<sup>1</sup>*Dip. di Fisica, Università di Roma "Tor Vergata", I-00133 Rome, Italy*

<sup>2</sup>*INFN, sez. Roma "Tor Vergata", I-00133 Rome, Italy*

<sup>3</sup>*Dip. di Fisica, Università di Roma "La Sapienza", I-00185 Rome, Italy*

<sup>4</sup>*INFN, sez. Roma, I-00185 Rome, Italy*

<sup>5</sup>*INFN, Laboratori Nazionali del Gran Sasso, 67010 Assergi (AQ), Italy*

<sup>6</sup>*Institute for Nuclear Research, MSP 03680 Kyiv, Ukraine*

<sup>7</sup>*Institute for Scintillation Materials, 61001 Kharkiv, Ukraine*

## Abstract.

Double beta processes in <sup>64</sup>Zn, <sup>70</sup>Zn, <sup>180</sup>W, and <sup>186</sup>W have been searched for with the help of low background ZnWO<sub>4</sub> crystal scintillators at the Gran Sasso National Laboratories of the INFN (Italy). The total measurement time exceeds ten thousand hours. New improved half-life ( $T_{1/2}$ ) limits on double electron capture and electron capture with positron emission in <sup>64</sup>Zn have been set. New  $T_{1/2}$  bounds were also set on different modes of  $2\beta$  processes in <sup>70</sup>Zn, <sup>180</sup>W, and <sup>186</sup>W. Future perspectives are considered.

## 1. INTRODUCTION

Experimental investigations in this field are concentrated mostly on  $2\beta^-$  decays, processes yielding the emission of two electrons. Results for double positron decay ( $2\beta^+$ ), electron capture with positron emission ( $\varepsilon\beta^+$ ), and capture of two electrons from atomic shells ( $2\varepsilon$ ) are much more modest. The most sensitive experiments give limits on the  $2\varepsilon$ ,  $\varepsilon\beta^+$  and  $2\beta^+$  processes at level of  $10^{17} - 10^{21}$  yr [1]. Reasons for this situation are: (1) lower energy releases in  $2\varepsilon$ ,  $\varepsilon\beta^+$  and  $2\beta^+$  processes in comparison with those in  $2\beta^-$  decay, that provide lower probabilities of the processes and difficult background suppression; (2) usually lower natural abundances of  $2\beta^+$  isotopes (which are typically lower than 1% with only few exceptions). Nevertheless, studies of neutrinoless  $2\varepsilon$  and  $\varepsilon\beta^+$  decays could help to explain the mechanism of neutrinoless  $2\beta^-$  decay (is it due to non-zero neutrino mass or to the right-handed admixtures in weak interactions) [2].

In Table 1 the potentially  $2\beta$  active nuclides present in ZnWO<sub>4</sub> crystals are listed.

## 2. MEASUREMENTS AND RESULTS

In Table 2 the ZnWO<sub>4</sub> crystal scintillators used in the present experiment are listed; they were produced from two crystal boules grown by the Czochralski method from ZnWO<sub>4</sub> compounds prepared from two batches of zinc oxide provided by different producers, and from the same tungsten oxide. The third scintillator ZWO-2a was cut from the ZWO-2 crystal.

To estimate the presence of naturally occurring radioactive isotopes, as well as some other elements important for growing the crystals, the ZnWO<sub>4</sub> samples were measured with the help of

**Table 1.** Potentially  $2\beta$  active nuclides present in  $\text{ZnWO}_4$  crystals.

Transition	Energy release (keV) [3]	Isotopic abundance (%) [4]	Decay channels	Number of nuclei in 100 g of $\text{ZnWO}_4$ crystal
$^{64}\text{Zn} \rightarrow ^{64}\text{Ni}$	1095.7(0.7)	48.268(0.321)	$2\varepsilon, \varepsilon\beta^+$	$9.28 \times 10^{22}$
$^{70}\text{Zn} \rightarrow ^{70}\text{Ge}$	998.5(2.2)	0.631(0.009)	$2\beta^-$	$1.21 \times 10^{21}$
$^{180}\text{W} \rightarrow ^{180}\text{Hf}$	144(4)	0.12(0.01)	$2\varepsilon$	$2.31 \times 10^{20}$
$^{186}\text{W} \rightarrow ^{186}\text{Os}$	489.9(1.4)	28.43(0.19)	$2\beta^-$	$5.47 \times 10^{22}$

**Table 2.**  $\text{ZnWO}_4$  crystal scintillators used in the present experiments.

Crystal scintillator	Size (mm)	Mass (g)
ZWO-1	$20 \times 19 \times 40$	117
ZWO-2	$\varnothing 44 \times 55$	699
ZWO-2a	$\varnothing 44 \times 14$	168

Inductively Coupled Plasma - Mass Spectrometry (ICP-MS, Agilent Technologies model 7500a) [5].

The  $\text{ZnWO}_4$  crystals were fixed inside a cavity of  $\varnothing 47 \times 59$  mm in the central part of a polystyrene light-guide 66 mm in diameter and 312 mm in length. The cavity was filled up with high purity silicone oil. The light-guide was optically connected on opposite sides by optical couplant to two low radioactivity 3" photomultipliers. The light-guide was wrapped by PTFE reflection tape. The detector has been installed deep underground in the low background DAMA/R&D set-up at the LNGS of the INFN (Italy). It was surrounded by Cu bricks and sealed in a low radioactive air-tight Cu box continuously flushed with high purity nitrogen gas (stored deeply underground for a long time) to avoid presence of residual environmental Radon. The copper box was surrounded by a passive shield made of 10 cm of high purity Cu, 15 cm of low radioactive lead, 1.5 mm of cadmium and 4/10 cm polyethylene/paraffin to reduce the external background. The whole shield has been closed inside a Plexiglas box, also continuously flushed by high purity nitrogen gas. An event-by-event data acquisition system accumulates the amplitude and the arrival time of the events. The sum of the signals from the PMTs was recorded with the sampling frequency of 20 MS/s over a time window of 100  $\mu\text{s}$  by a 8 bit transient digitizer (DC270 Acqiris).

The measurements were carried out in four runs (see Table 3 for details). First, the energy interval of data taking was chosen as 0.01 – 1 MeV with the aim to search for the neutrino accompanied double electron capture in  $^{64}\text{Zn}$ . Then the energy interval was changed to 0.05 – 4 MeV in order to search for other possible double beta processes in  $^{64}\text{Zn}$  (Run 2). Then the large ZWO-2 crystal was installed and the experiment was carried out in the same energy range (Run 3). Finally the ZWO-2 crystal was cleaved in two parts; one (ZWO-2a) was used in the Run 4 with the energy interval of data taking 0.01 – 1 MeV.

The energy scale and resolution of the  $\text{ZnWO}_4$  detectors have been measured with  $^{22}\text{Na}$ ,  $^{133}\text{Ba}$ ,  $^{137}\text{Cs}$ ,  $^{228}\text{Th}$  and  $^{241}\text{Am}$   $\gamma$  sources. In addition, the energy scale of the detectors was checked by using background  $\gamma$  lines of 609 keV of  $^{214}\text{Bi}$  (in all Runs), 1461 keV ( $^{40}\text{K}$ ), 1764 keV ( $^{214}\text{Bi}$ ), 2615 keV ( $^{208}\text{Tl}$ ) in Runs 2 and 3.

The knowledge of the radioactive contamination of the  $\text{ZnWO}_4$  crystals is necessary to describe the measured background spectra. The time-amplitude analysis, the pulse-shape discrimination, and the Monte Carlo simulation were applied to reconstruct the background spectra and to estimate the radioactive contamination of the  $\text{ZnWO}_4$  detectors. For details see ref. [5].

**Table 3.** Description of low background measurements with ZnWO<sub>4</sub> crystal scintillators. Times of measurements ( $t$ ), energy intervals of data taking ( $\Delta E$ ), energy resolutions at the 662 keV  $\gamma$  line of <sup>137</sup>Cs (FWHM), and background counting rates (BG) in different energy intervals are specified.

Run	Crystal scintillator	$t$ (h)	$\Delta E$ (MeV)	FWHM (%)	BG (counts/(day $\times$ keV $\times$ kg) in energy interval (MeV))			
					0.2–0.4	0.8–1.0	2.0–2.9	3.0–4.0
1	ZWO-1	1902	0.01–1	11.5	1.93(3)	0.27(1)		
2	ZWO-1	2906	0.05–4	12.6	1.71(2)	0.25(1)	0.0072(7)	0.0003(1)
3	ZWO-2	2130	0.05–4	14.6	1.07(1)	0.149(3)	0.0072(4)	0.00031(7)
4	ZWO-2a	3292	0.01–1	11.0	1.52(2)	0.211(7)		

The measured background spectra were fitted by the model built from the simulated distributions. Activities of U/Th daughters in the crystals were restricted taking into account the results of the time-amplitude and pulse-shape analyzes [5]. Activities of  $\beta$  active <sup>87</sup>Rb, <sup>113</sup>Cd, and <sup>115</sup>In were bounded taking into account the results of the ICP-MS analysis. The values of the <sup>40</sup>K, <sup>232</sup>Th and <sup>238</sup>U activities inside the PMTs were taken from [6] where the radioactive contamination of PMTs of the same model was measured. The peak in the spectrum of Run 3 at the energy  $1133 \pm 8$  keV cannot be explained by a contribution from external  $\gamma$  rays (the 1120 keV  $\gamma$  line of <sup>214</sup>Bi is not intense enough to provide the whole peak area). We assume the presence of <sup>65</sup>Zn ( $T_{1/2} = 244.26$  d,  $Q_{\beta} = 1351.9$  keV [7]) in the crystal to explain the peak. <sup>65</sup>Zn can be produced from <sup>64</sup>Zn by thermal neutrons (the cross section of <sup>64</sup>Zn to thermal neutrons is 0.76 barn [7]) or/and by cosmogenic activation. The result of the fit of the spectra of Run 2 and Run 3 in the energy region 0.1 – 2.9 MeV and the main components of the background as well as the summary of the radioactive contamination of the ZnWO<sub>4</sub> crystal scintillators (or limits on their activities) are given in [5].

There are no clear peculiarities in the measured energy spectra of the ZnWO<sub>4</sub> detectors, which can be interpreted as double beta decay of Zinc or Tungsten isotopes. Therefore only lower half-life limits can be set according to the formula:  $\lim T_{1/2} = N \cdot \eta \cdot t \cdot \ln 2 / \lim S$ , where  $N$  is the number of potentially  $2\beta$  unstable nuclei,  $\eta$  is the detection efficiency,  $t$  is the measuring time, and  $\lim S$  is the number of events of the effect searched for which can be excluded at a given confidence level (C.L.).

We have used different combinations of the accumulated data to reach maximal sensitivity to the sought double beta processes. The response functions of the ZnWO<sub>4</sub> detectors for the  $2\beta$  processes were simulated with the help of the GEANT4 code [8]. The initial kinematics of the particles emitted in the decays was generated with the DECAY0 event generator [9].

In ref. [5] the procedures followed to extract the limits on the various decay modes are described in details. All the half-life limits on  $2\beta$  decay processes in Zinc and Tungsten obtained in the present experiment are summarized in Table 4 where results of the most sensitive previous experiments are given for comparison.

The obtained bounds are well below the existing theoretical predictions [16, 17]; nevertheless most of the limits are near one order of magnitude higher than those established in previous experiments. It should be stressed that in contrast to a level of sensitivity obtained for double  $\beta^-$  decay ( $10^{23} - 10^{25}$  years in the best experiments), only two nuclei (<sup>40</sup>Ca and <sup>78</sup>Kr) among potentially  $2\varepsilon$ ,  $\varepsilon\beta^+$ ,  $2\beta^+$  active isotopes were investigated at the level of  $\sim 10^{21}$  yr. Moreover, the positive indication on the  $(2\nu + 0\nu)\varepsilon\beta^+$  decay of <sup>64</sup>Zn with  $T_{1/2} = (1.1 \pm 0.9) \times 10^{19}$  yr suggested in [14] is completely ruled out.

Further improvements in sensitivity can be reached by increasing the mass of the ZnWO<sub>4</sub> detector, suppression of external background and development of ZnWO<sub>4</sub> scintillators with lower level of radioactive contamination. High abundance of <sup>64</sup>Zn (48.3%) allows to build a large scale

**Table 4.** Half-life limits on  $2\beta$  processes in Zn and W isotopes.

Transition	Decay channel	Level of daughter nucleus	$T_{1/2}$ limit (yr) 90%(68%) C.L.	
			Present work	Previous results
$^{64}\text{Zn} \rightarrow ^{64}\text{Ni}$	$0\nu 2\varepsilon$	g.s.	$\geq 1.1(2.8) \times 10^{20}$	$\geq 0.7(1.0) \times 10^{18}$ [10] $\geq 3.4(5.5) \times 10^{18}$ [11]
	$0\nu \varepsilon \beta^+$	g.s.	$\geq 4.3(5.7) \times 10^{20}$	$\geq 2.8 \times 10^{16}$ [12] $\geq 2.4(3.6) \times 10^{18}$ [10] $\geq 1.3 \times 10^{20}$ [13] $\geq 2.2(6.1) \times 10^{20}$ [11]
	$2\nu \varepsilon \beta^+$	g.s.	$\geq 0.70(2.1) \times 10^{21}$	$= (1.1 \pm 0.9) \times 10^{19}$ [14] $\geq 4.3(8.9) \times 10^{18}$ [10] $\geq 1.3 \times 10^{20}$ [13] $\geq 2.1(7.4) \times 10^{20}$ [11]
$^{70}\text{Zn} \rightarrow ^{70}\text{Ge}$	$0\nu 2\beta^-$	g.s.	$\geq 1.8(3.0) \times 10^{19}$	$\geq 0.7(1.4) \times 10^{18}$ [10]
	$2\nu 2\beta^-$	g.s.	$\geq 2.3(4.0) \times 10^{17}$	$\geq 1.3(2.1) \times 10^{16}$ [10]
	$0\nu 2\beta^- \text{M1}$	g.s.	$\geq 1.0(1.4) \times 10^{18}$	
$^{180}\text{W} \rightarrow ^{180}\text{Hf}$	$0\nu 2\varepsilon$	g.s.	$\geq 0.86(1.2) \times 10^{18}$	$\geq 0.9(1.3) \times 10^{17}$ [15]
	$2\nu 2K$	g.s.	$\geq 6.6(9.4) \times 10^{17}$	$\geq 0.7(0.8) \times 10^{17}$ [15]
$^{186}\text{W} \rightarrow ^{186}\text{Os}$	$0\nu 2\beta^-$	g.s.	$\geq 2.1(4.2) \times 10^{20}$	$\geq 1.1(2.1) \times 10^{21}$ [15]
	$0\nu 2\beta^-$	$2^+$ (137.2 keV)	$\geq 2.1(4.2) \times 10^{20}$	$\geq 1.1(2.0) \times 10^{21}$ [15]
	$0\nu 2\beta^- \text{M1}$	g.s.	$\geq 5.8(8.6) \times 10^{19}$	$\geq 1.2(1.4) \times 10^{20}$ [15]
	$2\nu 2\beta^-$	g.s.	$\geq 2.3(2.8) \times 10^{19}$	$\geq 3.7(5.3) \times 10^{18}$ [15]
	$2\nu 2\beta^-$	$2^+$ (137.2 keV)	$\geq 1.8(3.6) \times 10^{20}$	$\geq 1.0(1.3) \times 10^{19}$ [15]

experiment without expensive isotopical enrichment. An experiment involving  $\approx 10$  tons of non-enriched crystals ( $9 \times 10^{27}$  nuclei of  $^{64}\text{Zn}$ ) could reach the half-life sensitivity  $\sim 3 \times 10^{28}$  yr (supposing zero background during ten years of measurements). Such a sensitivity could contribute to our understanding of the neutrino mass mechanism and right-handed currents in neutrinoless processes [2]. The two neutrino double electron capture should be surely observed: in accordance with theoretical expectations [16, 17],  $T_{1/2}$  for the  $2\nu 2\varepsilon$  process is predicted on the level of  $10^{25} - 10^{26}$  yr.

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