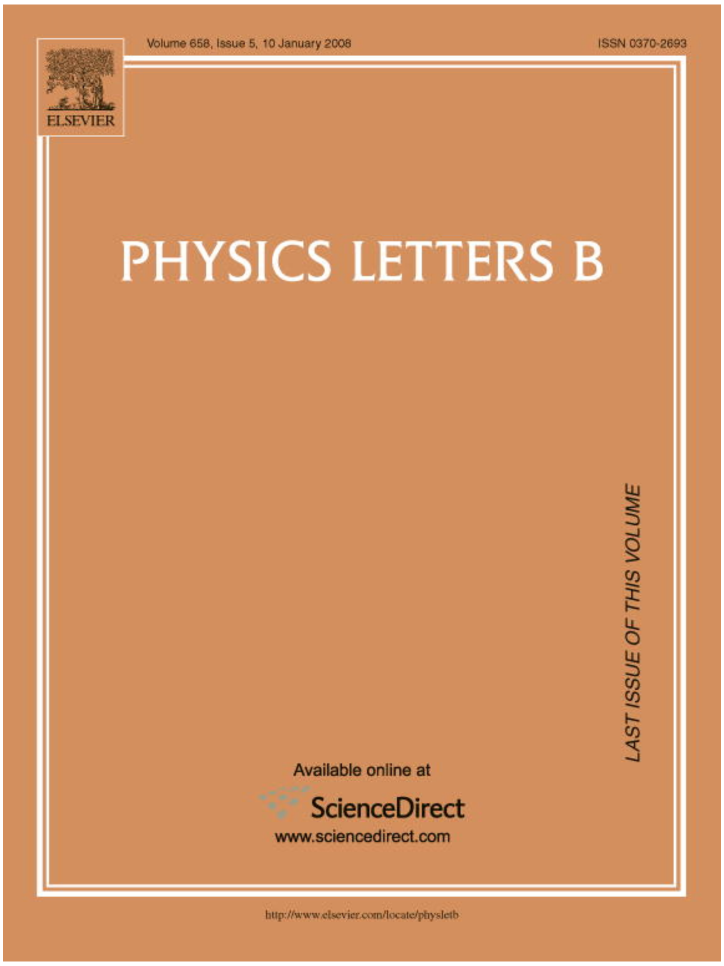
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Search for 2β processes in ^{64}Zn with the help of $ZnWO_4$ crystal scintillator

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Abstract

Double beta processes in ^{64}Zn were searched for with the help of a low background $ZnWO_4$ crystal scintillator (mass of 117 g) at the Gran Sasso National Laboratories of the INFN. Total time of measurements was 1902 h. New improved half-life limits on different modes of double electron capture and electron capture with positron emission were established as: $T_{1/2}^{2\nu 2K} \geqslant 6.2 \times 10^{18}$ yr, $T_{1/2}^{0\nu 2K} \geqslant 4.0 \times 10^{18}$ yr, $T_{1/2}^{0\nu 2\varepsilon} \geqslant 3.4 \times 10^{18}$ yr, $T_{1/2}^{2\nu\varepsilon\beta^+} \geqslant 2.1 \times 10^{20}$ yr, and $T_{1/2}^{0\nu\varepsilon\beta^+} \geqslant 2.2 \times 10^{20}$ yr, all at 90% C.L.

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Keywords: Double beta decay; ^{64}Zn; $ZnWO_4$ crystal scintillators

1. Introduction

Neutrinoless (0ν) double beta (2β) decay is one of the low-energy effects which are forbidden in the Standard Model (SM) because of violation of the lepton number on 2 units, but it is naturally expected in many SM extensions [1]. It offers complementary information to that given by neutrino oscillation experiments; in fact, while oscillation experiments are sensitive to the neutrinos mass difference, the $0\nu 2\beta$ decay experiments can allow to test the lepton number non-conservation, to establish the nature of the neutrino (that is, if it is a Majorana particle, $\nu = \bar{\nu}$, or a Dirac particle, $\nu \neq \bar{\nu}$) and to give the absolute scale of the effective neutrino mass.

Experimental investigations in this field are concentrated mostly on $2\beta^-$ decays, processes with emission of two electrons. Developments in the experimental techniques during the last two decades lead to observation of two neutrino (2ν) $2\beta^-$ decay in 10 isotopes with half-lives in the range of 10^{18}–10^{21} yr, and to sensitivities on $0\nu 2\beta^-$ decays up to 10^{23}–10^{25} yr. Searches for $2\beta^+$ decays, processes with emission of two positrons (or $\varepsilon\beta^+$, electron capture with positron emission; or 2ε, capture of two electrons from atomic shells) are not so popular [2]. Reasons for such a situation, in particular, are: (1) in general, lower energy releases in $2\beta^+$ decays in comparison with those in $2\beta^-$ decays, that results in higher expected $T_{1/2}$ values; (2) usually lower natural abundances of $2\beta^+$ isotopes (which very often are lower than 1% with only few exceptions), that restrict the number of nuclei available for investigations. Nevertheless, studies of $2\beta^+/\varepsilon\beta^+/2\varepsilon$ decays are important because they could help to distinguish the mech-

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anism of neutrinoless 2β decay (i.e., if it is due either to the non-zero neutrino mass or to the right-handed admixtures in weak interactions) [3].

In addition to ^{64}Zn, which is a main subject of this Letter and will be discussed in detail later, experimental searches for $2\beta^+/\varepsilon\beta^+/2\varepsilon$ processes were recently performed for ^{74}Se [4], ^{78}Kr [5], ^{106}Cd [6], ^{120}Te [7]. The obtained experimental $T_{1/2}$ limits depend on the nucleus, but usually they are on the level of 10^{18}–10^{19} yr. Full list of results till 2002 can be found in [8] and more recent achievements in [1,2].

^{64}Zn is one of a few exceptions among $2\beta^+$ nuclei having a big natural isotopic abundance of 48.268% [9]. This feature allows either to build a large scale experiment without isotopical enrichment or to make this procedure less expensive. The mass difference between ^{64}Zn and ^{64}Ni nuclei is 1095.7(0.7) keV [10] and, therefore, double electron capture (2ε), and electron capture with emission of positron ($\varepsilon\beta^+$) are energetically allowed [8].

An intriguing situation with this nucleus exists since 1995, when a possible experimental indication of the $\varepsilon\beta^+$ decay of ^{64}Zn with $T_{1/2}^{(0\nu+2\nu)\varepsilon\beta^+} = (1.1 \pm 0.9) \times 10^{19}$ yr was suggested in Ref. [11]. A $\varnothing$7.6 × 7.6 cm NaI(Tl) scintillator and a 25% efficiency HP Ge detector, operating in coincidence, were used in that experiment. The excess of $\approx$ 85 events in the 511 keV peak was observed with a zinc sample (mass of 350 g, 392 h of exposure on the sea level), while no effect was detected without a sample or with copper or iron blanks.

For long time, sensitivities of other experiments were not enough to confirm or disprove the result of Ref. [11]. A CdZnTe semiconductor detector was used to search for 2β decays of ^{64}Zn in underground measurements performed in the Gran Sasso National Laboratories ($\simeq$ 3600 m w.e.) over 1117 h in the COBRA experiment; however, the small mass of the detector (near 3 g) allowed to reach half-life limits at the level of 10^{16}–10^{17} yr [12]. These results were recently moderately improved with four CdZnTe crystals in the updated version of this experiment [13]. Low-background measurements with a small (4.5 g, 429 h of measurements) zinc tungstate ($ZnWO_4$) crystal scintillator was performed in the Solotvina Underground Laboratory ($\simeq$ 1000 m w.e.) [14]; despite the low mass of the detector, limits at a level of 10^{18} yr were set in the latter experiment for 2β processes in ^{64}Zn.

First results of another recent experiment were reported in some conferences in 2003 and recently appear in [15]. HP Ge detector 456 cm^3 and CsI(Tl) $\simeq$ 400 cm^3 were used in coincidence in measurements with 460 g Zn sample in the underground Cheong Pyung Laboratory ($\simeq$ 1000 m w.e.). Measurements during 375 h gave the limit on $\varepsilon\beta^+$ decay of ^{64}Zn as: $T_{1/2}^{(0\nu+2\nu)\varepsilon\beta^+} > 1.3 \times 10^{20}$ yr [15].

The aim of the present work is to search for double beta decays in ^{64}Zn with higher sensitivity with the help of large $ZnWO_4$ scintillators. The main properties of $ZnWO_4$ scintillators are: (i) density equal to 7.8 g/cm^3; (ii) light yield $\simeq$ 13% of that of NaI(Tl); (iii) refractive index equal to 2.1–2.2; (iv) emission maximum at 480 nm; (v) effective average decay time 24 μs. The material is non-hygroscopic and chemically inert; the melting point is at 1200 °C. Typical radiopurity of zinc tungstate crystals has been preliminarily investigated in [14]. As mentioned above, the present experiment has been carried out in the underground Gran Sasso National Laboratories of the INFN at a depth of $\simeq$ 3600 m w.e.; data were collected over a period of 1902 h.

2. The experimental set-up

A clear, slightly colored $ZnWO_4$ crystal (20 × 19 × 40 mm, mass of 117 g), produced from monocrystal grown by the Czochralski method, was used in our experiment. The $ZnWO_4$ crystal was fixed inside a cavity of $\varnothing$47 × 59 mm in central part of a polystyrene light-guide 66 mm in diameter and 312 mm in length. The cavity was filled up with high-purity silicon oil. The light-guide was optically connected on opposite sides by an optical couplant to two low radioactive EMI9265–B53/FL 3″ diameter photomultipliers (PMT). The light-guide was wrapped by the PTFE reflection tape. Such an assembling with use of oil allowed to increase the light transmission from the scintillator to PMTs and to improve the energy resolution of the detector [16].

The detector has been installed deep underground in the low background DAMA/R&D set-up at the Gran Sasso Laboratory. 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 the presence of residual environmental Radon. The Cu box has been 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 to 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 records the amplitude and the arrival time of events. Moreover, the sum of the signals from the PMTs was also recorded by a 1 GS/s 8 bit DC270 Transient Digitizer by Acqiris over a time window of 100 μs. To allow a good compromise to handle the data files and taking into account the slow scintillation decay of $ZnWO_4$, 20 MS/s sampling frequency was used during the data taking.

The time characteristics of $ZnWO_4$ scintillators under γ and α irradiation were studied in Ref. [14] with the help of a transient digitizer based on the 12 bit ADC (AD9022) operated at the sample rate of 20 MS/s. Three decay components $\tau_i \approx 0.7$, ≈ 7 and ≈ 25 μs with different amplitudes for γ rays and α particles were observed. These values offer the possibility to exploit in the production data the rejection of residual PMT noise[1] near a low energy threshold of $\simeq$ 15 keV (see Fig. 1). This procedure eliminates some part of scintillation signals near energy threshold. The energy dependence of the detection efficiency was determined with the help of ^{133}Ba, ^{137}Cs, ^{228}Th and ^{241}Am radioactive sources. The measured efficiency ranges

[1] Time characteristics of PMT noise were studied in the $ZnWO_4$ set-up in a special run removing the scintillation crystal from the light-guide; more than one million noise events were recorded.

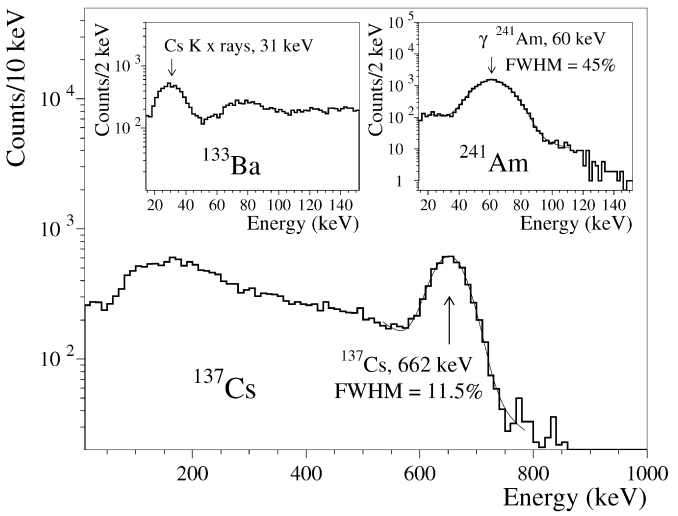

Fig. 1. Energy spectra measured by $ZnWO_4$ detector with ^{137}Cs (main part), and ^{133}Ba and ^{241}Am (insets) γ sources.

from about 0.30 at 15 keV up to about 0.65 at 30 keV. This has allowed us to investigate at least the higher energy part of the $2\nu 2K$ peak (see later) in spite of the relatively poor number of available photoelectrons/keV in the $ZnWO_4$ detector. For this purpose, both the optimal filter technique [17,18] and the mean time method [19] have been applied to the production data.

The energy scale and resolution of the $ZnWO_4$ detector for γ quanta were measured with ^{22}Na, ^{133}Ba, ^{137}Cs, ^{228}Th and ^{241}Am sources. The energy spectra accumulated with ^{133}Ba, ^{137}Cs and ^{241}Am are shown in Fig. 1. The energy dependence of the energy resolution can be fitted by the function: $\mathrm{FWHM}_\gamma(\mathrm{keV}) = \sqrt{270(30) + 7.37(35) \cdot E_\gamma}$, where E_γ is the energy of γ quanta in keV. For example, the width (FWHM) of the expected peak from the possible $2\nu 2K$ decay mode is $\simeq 22$ keV.

The data collection in current measurements was performed up to the energy of 1000 keV, that is related to the dynamical range of the digitizer.

3. Results and discussion

The energy spectrum measured with the $ZnWO_4$ crystal in the low background DAMA/R&D set-up over 1902 h, corrected for the energy dependence of detection efficiency, is presented in Fig. 2. The background level is consistent with the typical level of residual contamination in $ZnWO_4$ [14] and the set-up features, as confirmed by Monte Carlo simulation by using the GEANT4 [20] and DECAY0 [21] codes.

3.1. *Search for $\varepsilon\beta^+$ decays of ^{64}Zn*

The response functions of the $ZnWO_4$ detector for different 2β processes in ^{64}Zn were simulated with the help of the GEANT4 code [20]; the initial kinematics of the particles emitted in the decays was generated with the DECAY0 event generator [21]. The expected energy distributions are shown in Fig. 3.

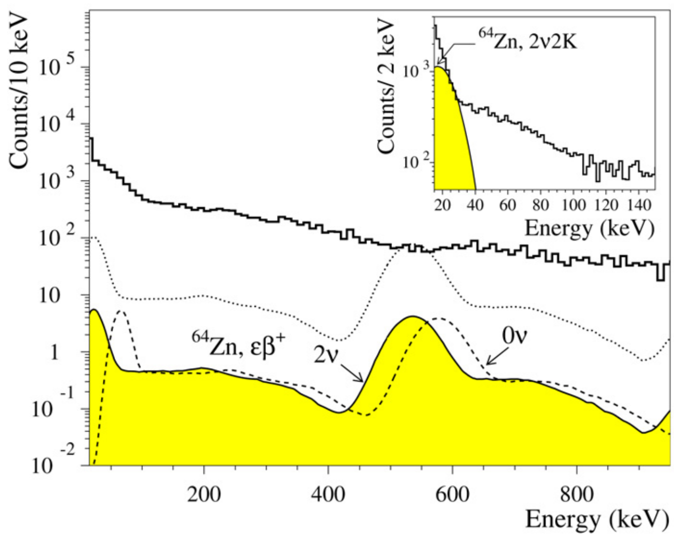

Fig. 2. The measured energy spectrum of the $ZnWO_4$ scintillation crystal (mass of 117 g, 1902 h of measurements)—corrected for the energy dependence of detection efficiency—together with the distributions for $\varepsilon\beta^+$ processes in ^{64}Zn excluded at 90% C.L. Energy spectrum of $2\nu\varepsilon\beta^+$ decay with $T_{1/2} = 1.1 \times 10^{19}$ yr (central value of positive indication in Ref. [11]) is also shown by dotted line. *In the Inset*: Low energy part of the spectrum together with the $2\nu 2K$ peak of ^{64}Zn with $T_{1/2} = 6.2 \times 10^{18}$ yr excluded at 90% C.L.

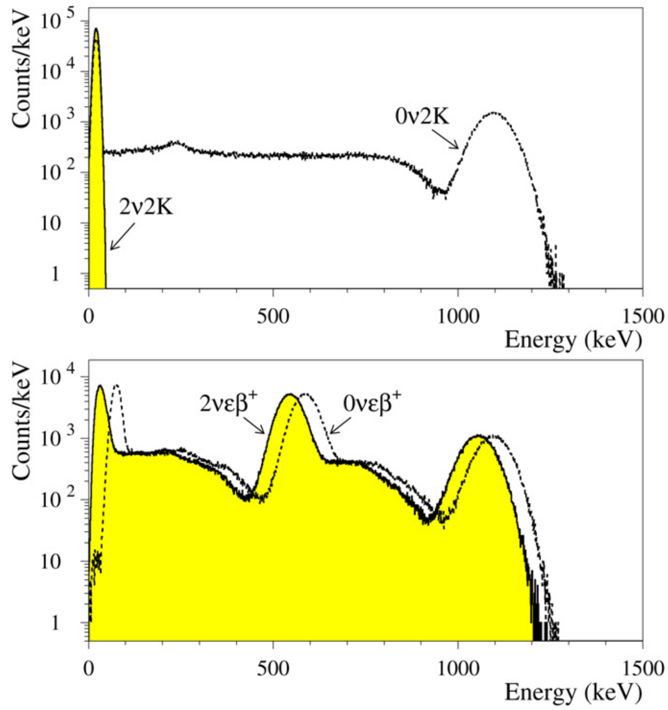

Fig. 3. Simulated response functions of the $ZnWO_4$ scintillator for different $\varepsilon\beta^+$ and 2ε decays in ^{64}Zn. One million of decays was simulated for each mode.

Comparing the simulated response functions with the experimental energy distribution accumulated with the $ZnWO_4$ crystal, we did not find in the latter one the peculiarities which can be unambiguously attributed to 2β processes in ^{64}Zn. Therefore only lower half-life limits can be expressed by the formula: $\lim T_{1/2} = N \cdot \eta \cdot t \cdot \ln 2 / \lim S$, where N is the number of potentially 2β unstable nuclei, η 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.).

Two approaches were used to estimate the value of $\lim S$ for the two neutrino mode of electron capture with positron emission ($2\nu\varepsilon\beta^+$) in ^{64}Zn. In the first one (the so-called 1σ approach), for such an estimation we used the statistical uncertainty of the number of events registered in the energy region of the expected peak, 470–600 keV, where 980 events were observed. Considering the number of ^{64}Zn nuclei in the crystal $N = 1.09 \times 10^{23}$, the measurement time $t = 1902$ h and the related efficiency $\eta = 0.41$, it gives the half-life limit $T_{1/2}^{2\nu\varepsilon\beta^+} \geqslant 2.2 \times 10^{20}$ yr. In the second approach, the experimental energy distribution was fitted in the energy interval 440–660 keV by the sum of the exponential function (representing background) and the expected model function for $2\nu\varepsilon\beta^+$ decay. The least squares fit ($\chi^2/n.d.f. = 17.3/20 = 0.87$) gives the total area of the effect as (-144 ± 117) counts which corresponds (in accordance with the Feldman–Cousins procedure [22]) to $\lim S = 80(22)$ counts at 90%(68%) C.L. Taking into account $\simeq 100\%$ registration efficiency for the whole curve in this case, one can calculate the half-life limit:

$$T_{1/2}^{2\nu\varepsilon\beta^+} \geqslant 2.1(7.4) \times 10^{20}\ \text{yr} \quad \text{at } 90\%(68\%)\ \text{C.L.}$$

In the same way the half-life bound on the neutrinoless mode was set as:

$$T_{1/2}^{0\nu\varepsilon\beta^+} \geqslant 2.2(6.1) \times 10^{20}\ \text{yr} \quad \text{at } 90\%(68\%)\ \text{C.L.}$$

The energy distributions expected for the $2\nu\varepsilon\beta^+$ and $0\nu\varepsilon\beta^+$ decay of ^{64}Zn, excluded at 90% C.L., are shown in Fig. 2. There is also shown the $2\nu\varepsilon\beta^+$ spectrum, which should be expected in our experiment when $T_{1/2} = 1.1 \times 10^{19}$ yr (which is the central value of the positive indication by Ref. [11]) is assumed (dotted line). In this case all the events observed in our experiment in the region of the peak at $\simeq 550$ keV have to be ascribed only to $2\nu\varepsilon\beta^+$ decay of ^{64}Zn, without contribution from the background; however, such an additional contribution exists, as can be derived considering the left and the right sides of the $\simeq 550$ keV peak. Therefore, the positive indication with $T_{1/2} = 1.1 \times 10^{19}$ yr is excluded with high confidence by the present experimental data. This confirms and improves the result recently appeared in [15].

3.2. *Double electron capture in ^{64}Zn*

For the 2ν double electron capture in ^{64}Zn from the K shell, the total energy released in the detector is equal to $2E_K = 16.7$ keV (where $E_K = 8.3$ keV is the binding energy of electrons on the K shell of nickel atoms). Detection of such a little energy deposit requires rather low energy threshold. In our measurements the energy threshold for acquisition was enough low (see above) to observe at least the higher energy part of the $2\nu2K$ peak.

To set a limit on the $2\nu2K$ decay of ^{64}Zn, taking into account the proximity of the energy threshold and the contribution from remaining PMT noise (consider, as mentioned, the modest number of available photoelectrons/keV), we just use a very simple and conservative requirement: the theoretical energy distribution should not exceed the experimental one in any energy interval above energy threshold, including error bars in the experimental values. In this way the limit on the peak area is 2662(2603) counts at 90(68)% C.L. Using this value for the peak area, we conservatively give only the half-life limit on the process as:

$$T_{1/2}^{2\nu2K} \geqslant 6.2(6.3) \times 10^{18}\ \text{yr} \quad \text{at } 90\%(68\%)\ \text{C.L.}$$

As regards the neutrinoless double electron captures, in the following we suppose that—in addition to de-excitation processes in atomic shells—a further energy release in the decay is taken away by (bremsstrahlung) γ quantum. In this case, for the 0ν double capture from the K shell, a peak at the energy of 17 keV is expected if the emitted γ quantum escapes the detector without interaction (see Fig. 3). In this case the following limit is obtained:

$$T_{1/2}^{0\nu2K} \geqslant 4.0(4.1) \times 10^{18}\ \text{yr} \quad \text{at } 90(68)\%\ \text{C.L.}$$

For the 0ν electron captures from the K and L (or $2L$) shells, the low energy peak will be below the energy threshold in the present experiment. However, we can use the high energy part of the spectrum near 1096 keV, when all emitted particles (high-energy γ and particles from de-excitations in atomic shells) are absorbed in the detector. While the energy scale of the current experiment was restricted by a value of 1000 keV, it nevertheless gives the possibility to observe the left part of the expected peak (see Fig. 2). $2K$, KL, $2L$ (and other) modes are not energetically resolved in the high energy region because of the finite energy resolution of the detector. Fitting of the measured spectrum in the energy interval 700–1000 keV by the sum of the exponential function (representing the background) and of the expected behaviour gives the following general limit:

$$T_{1/2}^{0\nu2\varepsilon} \geqslant 3.4(5.5) \times 10^{18}\ \text{yr} \quad \text{at } 90\%(68\%)\ \text{C.L.}$$

All obtained limits are summarised in Table 1 where results of the most sensitive previous experiments are listed too. The limits established in the present work are more stringent than those previously obtained by some of the authors in the experiment with small $ZnWO_4$ crystal at Solotvina Laboratory [14], and are better (up to four orders of magnitude) than limits from other experiments [11–13,15].

New measurements are in progress with an increased energy scale. As a next step, we plan to install a larger crystal of 0.7 kg mass which could allow to improve the sensitivity of the experiment to $\varepsilon\beta^+$ decay of ^{64}Zn up to the level of 10^{21} yr. Following this, a $ZnWO_4$ crystal with much better light output will be installed to more carefully explore the low energy part of the $ZnWO_4$ energy spectrum, where the $2\nu2K$ process in ^{64}Zn is expected. Further improvement of the experiment could be reached by suppression of background with longer light-guides produced from high-purity material (quartz, or $PbWO_4$ scintillator as was proposed in [23]) to suitably shield a $ZnWO_4$ detector from external γ quanta from PMTs. In addition, the

Table 1
Half-life limits on 2β processes in the decay ^{64}Zn → ^{64}Ni at 90% C.L.

Decay channel	Experimental $T_{1/2}$, yr	
	Present work	Previous results
$0\nu\varepsilon\beta^+$	$> 2.2 \times 10^{20}$	$> 2.8 \times 10^{16}$ [12]
		$> 2.4 \times 10^{18}$ [14]
		$> 1.3 \times 10^{20}$ [15]
$2\nu\varepsilon\beta^+$	$> 2.1 \times 10^{20}$	$= (1.1 \pm 0.9) \times 10^{19}$ [11]
		$> 4.3 \times 10^{18}$ [14]
		$> 1.3 \times 10^{20}$ [15]
$0\nu 2K$	$> 4.0 \times 10^{18}$	$> 1.2 \times 10^{17}$ [13]
$2\nu 2K$	$> 6.2 \times 10^{18}$	$> 6.0 \times 10^{16}$ [12]
$0\nu 2\varepsilon$	$> 3.4 \times 10^{18}$	$> 7.0 \times 10^{17}$ [14]

search for double beta decays of ^{70}Zn, ^{180}W and ^{186}W can be realized as a by-product of the experiment.

4. Conclusions

A low background experiment to search for double electron capture and electron capture with positron emission in ^{64}Zn was carried out over 1902 h in the underground Gran Sasso National Laboratories of INFN by using a $ZnWO_4$ scintillation detector with mass of 117 g.

New improved limits were set for different modes of double beta decay of ^{64}Zn by analyzing the energy distribution measured with the $ZnWO_4$ detector. In particular, the $2\nu 2K$ and $2\nu\varepsilon\beta^+$ processes were restricted at the level of $T_{1/2}^{2\nu 2K} \geqslant 6.2 \times 10^{18}$ yr and $T_{1/2}^{2\nu\varepsilon\beta^+} \geqslant 2.1 \times 10^{20}$ yr at 90% C.L., respectively. The positive indication on the $(2\nu + 0\nu)\varepsilon\beta^+$ decay of ^{64}Zn suggested in [11] is discarded by the present experiment. This confirms and improves the result recently appeared in [15].

Measurements are in progress with a higher energy scale. As a next step, we plan to install a larger crystal with mass of 0.7 kg. Following this, also measurements by using a $ZnWO_4$ scintillator with higher light output are foreseen to improve sensitivity to $2\nu 2K$ process in ^{64}Zn.

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