Search for the β decay of ^{96}Zr

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Abstract

^{96}Zr and ^{48}Ca are unique among double- β decay candidate nuclides in that they may also undergo single- β decay. In the case of ^{96}Zr , the single- β decay mode is dominated by the fourth-forbidden β decay with a 119 keV Q value. A search was conducted for the β decay of ^{96}Zr by observing the decay of the daughter ^{96}Nb nucleus. Two coaxial high-purity germanium detectors were used in coincidence to detect the γ -ray cascade produced by the daughter nucleus as it de-excited to the ground state. The experiment was carried out at the Kimballton Underground Research Facility and produced 685.7 days of data with a 17.91 g enriched sample. No counts were seen above background, producing a limit of $T_{1/2} > 2.4 \times 10^{19}$ yr. This is the first experimental search that is able to discern between the β decay and double- β decay to an excited state of ^{96}Zr .

Keywords: Beta decay, ^{96}Zr , Gamma-ray spectroscopy, Gamma coincidence, Underground detector

1. Introduction

Two-neutrino double- β ($2\nu\beta\beta$) decay is a second order nuclear decay that has been observed in 11 nuclides. Among these 11, ^{96}Zr is unique, along with ^{48}Ca , in that the single β decay is energetically allowed, but highly suppressed. The 0^+ ^{96}Zr nucleus must undergo a sixth forbidden β decay to the 6^+ ground state or a fourth forbidden β decay to the first two excited states, 5^+ and 4^+ , of ^{96}Nb . Reference [1] theoretically estimated that the decay is dominated by the transition to the 5^+ state with a half-life of 2.4×10^{20} yr. This estimate is ten times larger than the half-life of the $2\nu\beta\beta$ decay to the ground state of ^{96}Mo measured by the NEMO-3 collaboration: $T_{1/2} = [2.35 \pm 0.14(\text{syst}) \pm 0.16(\text{stat})] \times 10^{19}$ yr [2].

Figure 1: The single- β decay of ^{96}Zr and its daughter, ^{96}Nb . The figure is not to scale and all energy values are given in keV.

31 A current interest is measuring $2\nu\beta\beta$ decay to excited nuclear states.
 32 Measurement of this decay mode may provide additional information on the
 33 nuclear structure of the decay and aid in calculations of the nuclear matrix
 34 element. Furthermore, decays to an excited state could be used as a consis-
 35 tency test for $0\nu\beta\beta$ in ton-scale experiments using one isotope [3]. The most
 36 likely excited state $2\nu\beta\beta$ decay is to the first excited 0_1^+ state. For ^{96}Zr this
 37 has an expected half-life comparable to that of the β decay mode [4]. The
 38 β decay daughter, ^{96}Nb decays with a half-life of 23.35 hours and emits a
 39 cascade of γ rays, as is shown in Fig. 1.

40 2. Experimental Method

41 The apparatus was initially constructed to search for $2\nu\beta\beta$ decays to
 42 excited final states. The results of the search for the $2\nu\beta\beta$ decay of ^{96}Zr to
 43 excited states is given in Ref. [5], and a complete description of the detector
 44 system is given in Ref. [6]. Two γ rays from the γ -ray cascade are detected
 45 in coincidence by two high-purity germanium (HPGe) detectors in order to
 46 minimize the experimental background. Previous limits on the β and $\beta\beta$
 47 decay to excited states used a single HPGe detector and were focused on the
 48 detection of the single 778.2 keV γ ray [7, 8]. These works produced high
 49 quality limits, but as this γ ray could be produced from either the β decay or
 50 the $\beta\beta$ decay to the first excited 0_1^+ state, the technique is inherently limited
 51 as it cannot distinguish between the two decay modes.

Figure 2: (Color online) Diagram of the detector system. The copper plates are not shown and are located between the NaI annulus and lead shielding.

2.1. Detector apparatus

Two coaxial HPGe detectors sandwich a disk-shaped sample. Both germanium crystals are 88 mm in diameter and 50 mm in length. The HPGe detectors are surrounded by a NaI annulus, which is used as an active veto and Compton suppression shield. Additionally, two plastic scintillator end caps are positioned on each side of the annulus to provide additional veto power. The entire apparatus is surrounded by two layers of passive shielding: 0.75" oxygen-free high conductivity copper and 6" of lead. The experimental apparatus is housed in the Kimballton Underground Research Facility (KURF), which provides 1450 m of water equivalent overburden [9]. A diagram of this apparatus is shown in Fig. 2.

Five signals are recorded for each generation of the master trigger: the pulse height of each HPGe detector, the timing between the HPGe detectors, and two copies of the veto timing. Every event originating in a HPGe detector is recorded and the coincidence criteria is enforced in software. The HPGe detector readout is calibrated to energy approximately every four days using at least four naturally occurring background peaks. Each run is examined for gain shifts, and runs exhibiting such behavior are excluded from the analysis. The long term stability of the HPGe detectors was monitored by investigating the detector's count rate, energy resolution, and efficiency.

Table 1: The three most probably decay sequences for the 5^+ state of ^{96}Mo . The branching ratio f_b and the energy of the three de-excitation γ rays is given for each sequence.

Decay sequence	f_b	γ_1 [keV]	γ_2 [keV]	γ_3 [keV]
$5_0^+ \rightarrow 4_1^+ \rightarrow 2_0^+ \rightarrow 0_0^+$	51.5%	568.9	1091.3	778.2
$5_0^+ \rightarrow 3_0^+ \rightarrow 2_0^+ \rightarrow 0_0^+$	18.8%	460.0	1200.0	778.2
$5_0^+ \rightarrow 4_0^+ \rightarrow 2_0^+ \rightarrow 0_0^+$	11.3%	810.8	849.9	778.2

2.2. Enriched ^{96}Zr sample

Enriched $^{96}\text{ZrO}_2$ samples were leased from Oak Ridge National Laboratory, as the small natural abundance of ^{96}Zr , 2.8%, can limit the experimental sensitivity. A 7.2835 g sample enriched to 91.4% ^{96}Zr and a 26.9685 g sample enriched to 64.2% ^{96}Zr were placed in an acrylic container of diameter of 69.95 mm and thickness of 10.22 mm. The two samples together total 17.914 g of ^{96}Zr . 685.7 days (1.877 yr) of data were produced with this enriched sample in place.

3. Analysis procedure

The transitions in ^{96}Nb are below the detection threshold of the current experiment. ^{96}Nb decays with a 23.35 hour half-life to ^{96}Mo , with a branching ratio of 96.7% to the 5^+ state. The 5^+ state then de-excites, producing a γ -ray cascade ideal for detection by the two-coaxial HPGe apparatus. Detection of the de-excitation γ rays from the 5^+ state would provide a direct measurement of ^{96}Zr 's β decay. The three most probable decay modes are given in Table 1. These decay modes represent 81.6% of all possible decays, with no single remaining decay having a branching ratio $> 4.5\%$. All of these decays contain three coincident γ rays, which differentiates the analysis from that of $\beta\beta$ decay to excited states, where the signal region involves only two coincident γ rays.

3.1. Event selection

The region of interest (ROI) for β decay was chosen very carefully in order to maximize the signal-to-noise ratio while minimizing systematic uncertainties involved in calculating the detector efficiency. The result is a search for events where two of the three γ rays are detected in coincidence, with the third escaping the HPGe detectors. This is referred to as a two γ -ray event.

No cuts are made on the NaI annulus, as the third γ ray may interact in the annulus. The background was found to be sufficiently low without using the NaI veto due to the coincidence between the two HPGe detectors.

A second search procedure was conducted for events where two γ rays have complete energy deposition in one HPGe detector and the third γ ray is detected in coincidence. This is referred to as a three γ -ray event. In a three γ -ray events, the NaI is used in anticoincidence, as full energy deposition occurs in the HPGe detectors and leaves no signal in the NaI annulus. Although the three- γ ray event ROI has a smaller efficiency than two γ -ray events, by a factor of 5-6, the summing of the two γ rays will result in higher energy event with a corresponding lower background.

Candidate events must match the energy of the ROI, within $\pm 2\sigma$ of the detector resolution, and the expected background was extrapolated from the data. This is the same analysis procedure detailed in Ref. [5] for the $2\nu\beta\beta$ decay to excited states. The resulting histograms are shown in Fig. 3 for the decay mode with the highest branching ratio, $5_0^+ \rightarrow 4_1^+ \rightarrow 2_0^+ \rightarrow 0_0^+$. The top two histograms illustrate the search for two γ -ray events, while the bottom three histograms show the search for three γ -ray events.

The results of this analysis are detailed in Table 2. The analysis is performed for the three decay modes detailed in Table 1. Each decay mode produces three γ rays, resulting in three different two γ -ray events. There are three additional possible three γ -ray events, giving a total of six possible coincident regions of interest per decay mode. Of these, three were found to be unfit for analysis and are not included. The 568.9-778.2 keV coincidence was contaminated by the 569.8-1063.7 keV γ -ray coincidence resulting from neutron scattering on lead. The 460.0-778.2 keV coincidence was contaminated by the 463.0-911.2 keV γ -ray coincidence from ^{228}Th . Finally, the $1200.0 + 778.2 = 1978.2$ keV region of interest was excluded because it appears close to the ADC cutoff energy, where nonlinearities in the calibration could possibly occur.

3.2. Efficiency calculation

The coincidence efficiency of the apparatus was measured using a ^{102}Rh source. This source was chosen because it exhibits both the $0^+ \rightarrow 2^+ \rightarrow 0^+$ and $2^+ \rightarrow 2^+ \rightarrow 0^+$ decay mode. The efficiency was measured as a function of radius for eight points between $0 \leq r \leq 4.5$ cm and in four different directions (up/down/left/right). For the measurement of the $2\nu\beta\beta$ decay, corrections were made to account for the energy of the γ rays, the target

Figure 3: The ROI histograms for a subset of the ^{96}Zr β -decay coincidences. Histograms are shown for the $5_0^+ \rightarrow 4_1^+ \rightarrow 2_0^+ \rightarrow 0_0^+$ decay. The minimum detectable signal at the 90% C.L. is illustrated by the red curve. (a-c) are all in coincidence with 1091.3 keV and show the 568.9, 778.2, and 1347.1 keV ROIs respectively. Histogram (d) is in coincidence with 568.9 keV and shows the 1869.5 keV ROI and (e) is in coincidence with 778.2 keV and shows the 1660.2 keV ROI. The data acquisition time was 1.877 yr.

Table 2: The results of the search for ^{96}Zr 's β decay are summarized. For each decay sequence and coincident pair the effective efficiency ϵ_{eff} and its associated systematic error σ_{syst} , number of observed counts in the ROI N_{obs} , and the expected background N_{bkgd} are given.

Decay sequence	Given γ [keV]	Second γ [keV]	ϵ_{eff} [%]	σ_{syst}	N_{obs}	N_{bkgd}
$5_0^+ \rightarrow 4_1^+ \rightarrow 2_0^+ \rightarrow 0_0^+$	1091.3	568.9	0.0856	7.5%	3	1.35
	1091.3	778.2	0.0660	7.6%	1	1.10
	1091.3	1347.1 ¹	0.0127	7.7%	0	0.24
	568.9	1869.5 ¹	0.0141	7.6%	0	0.36
	778.2	1660.2 ¹	0.0141	7.7%	0	0.18
$5_0^+ \rightarrow 3_0^+ \rightarrow 2_0^+ \rightarrow 0_0^+$	460.0	1200.0	0.0939	7.5%	1	1.99
	778.2	1200.0	0.0598	7.6%	1	0.77
	778.2	1660.0 ¹	0.0149	7.6%	0	0.15
	1200.0	1238.2 ¹	0.0137	7.7%	1	0.28
$5_0^+ \rightarrow 4_0^+ \rightarrow 2_0^+ \rightarrow 0_0^+$	778.2	849.9	0.0833	7.5%	2	1.40
	778.2	810.8	0.0752	7.5%	3	1.57
	849.9	810.8	0.0801	7.6%	0	1.25
	778.2	1660.7 ¹	0.0131	7.6%	0	0.21
	849.9	1589.0 ¹	0.0120	7.7%	0	0.09
	810.8	1628.1 ¹	0.0130	7.7%	1	0.14

¹Sum of two coincident γ rays (a three γ -ray event)

Figure 4: (Color online) Efficiency as a function of energy for both detectors. The simulation data were produced using GEANT4. The functional fit is shown by the black curve.

135 attenuation, the detector separation, the spatial extent of the target, and
 136 the angular distribution of the γ rays.

137 When extending these efficiency measurements with two coincident γ rays
 138 to three coincident γ rays, the corrections become difficult to disentangle.
 139 These issues are resolved by using GEANT4 simulations of the efficiency nor-
 140 malized to the ^{102}Rh efficiency. All of the efficiency corrections for the $2\nu\beta\beta$
 141 decay mode were performed using GEANT4 to extreme accuracy [5]. The
 142 only correction previously done using experimental data and now done using
 143 GEANT4 is the energy dependence of the efficiency, which is shown in Fig. 4
 144 to have excellent agreement with measurements performed using a ^{152}Eu
 145 source. The correction factor was calculated using both the measured and
 146 simulated efficiency curves for all possible γ -ray pairs resulting from ^{96}Zr 's
 147 β decay. The correction factor differed by $< 1.4\%$ between the simulated
 148 and measured values, with the higher discrepancy occurring for the higher
 149 energy γ rays. This difference was added as a systematic uncertainty for the
 150 β -decay efficiency calculation.

151 The regions of interest for β decay are still coincidences where both de-
 152 tectors are triggered, meaning the trigger efficiency is identical for the inves-
 153 tigated ROI and the efficiency measurements. The interaction probability of
 154 the third γ ray has the largest uncertainty. The angular distribution, inter-
 155 action cross section of γ ray, and detector geometry are all well known, so
 156 the simulation is well equipped to deal with the third γ ray. The uncertainty

Figure 5: The efficiency as a function of position for ^{96}Zr 's β decay. The decay contains three coincident γ rays (568.9, 1091.3, and 778.2 keV). The efficiency is shown for events given observation of a 1091.3 keV γ ray in one detector. The other detector will see peaks at 568.9 and 778.2 keV (two γ ray events), and $568.9 + 778.2 = 1347.1$ keV (three γ -ray event).

on the single HPGe detector efficiency in the GEANT4 simulation (5.5%) is added into the error budget as an additional uncertainty. This is estimated from the maximum discrepancy between the GEANT4 efficiency simulations and the measured efficiency in a single HPGe.

Results of this simulation are shown by the efficiency curves in Fig. 5. Each of the ROIs in Table 2 has its own efficiency curve, only three of which are shown here. The effective efficiency ϵ_{eff} in Table 2 is the efficiency resulting from averaging $\epsilon(r)$ over the ^{96}Zr sample size.

A summary of systematic errors is shown in Table 3. The largest source of error (3.1%) comes from the calibrated ^{137}Cs source that was used to determine the total detector efficiency. The second largest effect (2.5%) comes from the uncertainty in the detector and source placement. Small asymmetries discovered when measuring the radial efficiency and were estimated to contribute a 2.4% uncertainty in the overall efficiency. The final uncertainty, the statistical uncertainty of the GEANT4 simulations, results from fitting the efficiency curve in Fig. 5.

3.3. Limit setting

The results of the individual ROIs may be summed to form one statistically significant result. The number of observed events exceeds the back-

Table 3: The systematic error budget for the β decay of ^{96}Zr . These uncertainties are for the 1091.3-568.9 keV coincidence. Note that uncertainty on the energy dependence and statistical uncertainty vary for the different transitions.

Uncertainty contribution	Percent error
Calibration of ^{137}Cs source	3.1 %
Energy dependence correction factor	0.4 %
Attenuation correction factor	0.2 %
Z dependence correction factor	0.15 %
Detector and source geometry	3.0 %
Non-symmetrical efficiency curve	2.4 %
Dead time	0.15 %
Uncertainty in ^{102}Rh half-life	0.82 %
Angular distributions	0.30 %
Total efficiency of HPGe in GEANT4	5.5 %
Energy dependence of efficiency in GEANT4	0.92 %
Statistical uncertainty of GEANT4	0.61 %
Total uncertainty	7.5 %

ground prediction, but is statistically consistent with the expected back-
ground. Lower limits were set using the formula

$$T_{1/2} > \frac{\ln 2 N_0 t f_b \epsilon_{eff}}{N_d}, \quad (1)$$

where N_0 is the number of nuclei, t is the exposure time, f_b is the branching ratio, ϵ_{eff} is the efficiency, and N_d is a statistical factor representing the number of counts above background to which the experiment is sensitive. N_d was calculated using the method of Feldman and Cousins [10] for a Poisson process and a 90% confidence level. The systematic uncertainty on the efficiency is included in limit setting at the 90% confidence level.

This analysis is shown in Table 4. The final result is a combined limit of $T_{1/2}(\beta(^{96}\text{Zr})) > 2.4 \times 10^{19}$ yr. Noticeably, the same limit may be produced using only the most intense $5_0^+ \rightarrow 4_1^+ \rightarrow 2_0^+ \rightarrow 0_0^+$ transition. Additional transitions increase the background, thus reducing the apparatus's total sensitivity, at the same rate at which they increase the total efficiency. The limit produced using only the search for two γ -ray events is a very comparable 2.1×10^{19} yr.

Table 4: Half-life limits for ^{96}Zr 's β decay from the combined statistical analysis at the 90% confidence level.

Decay sequence	Two- γ [yr]	Three- γ [yr]	Combined [yr]
$5_0^+ \rightarrow 4_1^+ \rightarrow 2_0^+ \rightarrow 0_0^+$	1.6×10^{19}	1.6×10^{19}	2.4×10^{19}
$5_0^+ \rightarrow 3_0^+ \rightarrow 2_0^+ \rightarrow 0_0^+$	1.1×10^{19}	0.18×10^{19}	1.0×10^{19}
$5_0^+ \rightarrow 4_0^+ \rightarrow 2_0^+ \rightarrow 0_0^+$	0.61×10^{19}	0.14×10^{19}	0.66×10^{19}
Combined	2.1×10^{19}	0.94×10^{19}	2.4×10^{19}

191 4. Conclusions

192 Unfortunately, this limit is not sufficient to test the theoretical limit of
193 2.4×10^{20} yr by Ref. [1]. Furthermore, the present limit is less than the previ-
194 ous limit of 3.8×10^{19} [7]. The previous work used a single detector to search
195 for the 778.2 keV γ ray. This technique results in a higher efficiency, but with
196 an increased background. As the present experiment covers a larger solid an-
197 gle, the coincidence summing of the three coincident γ rays becomes a large
198 factor and noticeably decreases the efficiency compared to a single detector
199 with half the solid angle. Although the technique of Ref. [7] is sufficient for
200 limit setting, any observation would require detection of coincident γ rays
201 to distinguish the β decay from the $2\nu\beta\beta$ decay to excited states. As such,
202 the present experiment has a much higher discovery potential even though
203 the produced limits may not be as competitive. The background reduction
204 provided by the $\gamma - \gamma$ coincidence has also been shown in the present work
205 and will be necessary in any future searches.

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212 References

- 213 [1] H. Heiskanen, M. T. Mustonen, and J. Suhonen, J. Phys. G **34**, 837
214 (2007).

- 215 [2] J. Argyrlades *et al.* (NEMO-3 Collaboration), Nucl. Phys. A **847**, 168
216 (2010).
- 217 [3] M. Duerr, M. Lindner, and K. Zuber, Phys. Rev. D **84**, 093004 (2011).
- 218 [4] Y. Ren and Z. Ren, Phys. Rev. C **89**, 064603 (2014).
- 219 [5] S. W. Finch and W. Tornow, accepted to Phys. Rev. C (2015).
- 220 [6] M. F. Kidd, J. H. Esterline, S. W. Finch, and W. Tornow, Phys. Rev. C
221 **90**, 055501 (2014).
- 222 [7] C. Arpesella, A. S. Barabash, E. Bellotti, C. Brofferio, E. Fiorini,
223 P. P. Sverzellati, and V. I. Umatov, Europhys. Lett. **27**, 29 (1994).
- 224 [8] A. S. Barabash, R. Gurriarán, F. Hubert, J. L. Reyss, J. Suhonen, and
225 V. I. Umatov, J. Phys. G **22**, 487 (1996).
- 226 [9] P. Finnerty *et al.*, Nucl. Instrum. Methods A **642**, 65 (2011).
- 227 [10] G. J. Feldman and R. D. Cousins, Phys. Rev. D **57**, 3873 (1998).