# First limit on neutrinoless quadruple $\beta$ decay of <sup>150</sup>Nd to the 0<sup>+</sup><sub>1</sub> state of <sup>150</sup>Gd

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**Background:** Observation of lepton number violation via detection of neutrinoless double  $\beta$  decay requires that the neutrino be a Majorana particle. If the neutrino is a Dirac particle, a potential lepton-number violating process is neutrinoless quadruple  $\beta$  decay. Only a few nuclei can undergo neutrinoless quadruple  $\beta$  decay; one of these nuclei is <sup>150</sup>Nd.

**Purpose:** This study yields the first half-life limit of the neutrinoless quadruple  $\beta$  decay to the excited  $0_1^+$  state of <sup>150</sup>Gd.

**Methods:** We searched for neutrinoless quadruple  $\beta$  decay events to excited final states of <sup>150</sup>Gd by detecting the deexcitation  $\gamma$  rays of the daughter nucleus in coincidence. These  $\gamma$  rays have energies of 569.031 and 638.050 keV, and are emitted in coincidence through a  $0^+_1 \rightarrow 2^+_1 \rightarrow 0^+_{gs}$  transition.

**Results:** The enriched Nd<sub>2</sub>O<sub>3</sub> sample consisted of  $40.33 \pm 0.02$  g<sup>150</sup>Nd and was observed for 642.8 days at the Kimballton Underground Research Facility. A half-life limit for the decay to the 0<sup>+</sup><sub>1</sub> state of <sup>150</sup>Gd was found to be  $T_{1/2} > 1.76 \times 10^{20}$  years (90% CL).

**Conclusions:** We report the first search for this decay to excited final states. Though the predicted half-life of this decay is many orders of magnitudes larger, constraining this value experimentally is vital to check for potential enhancements to the decay rate.

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# I. INTRODUCTION

One major result that would come from the detection of neutrinoless double- $\beta$  ( $0\nu\beta\beta$ ) decay is the observation of lepton number violation (LNV). For  $0\nu\beta\beta$  decay to proceed, the neutrino must be its own antiparticle, a Majorana particle. If the neutrino is a Dirac particle,  $0\nu\beta\beta$  decay will not proceed. However, as stated by Heeck and Rodejohann [1], the Majorana nature of the neutrino is not required for LNV. One possible allowed decay with Dirac neutrinos that would demonstrate LNV is neutrinoless quadruple  $\beta$  decay ( $0\nu4\beta$ ),

$$(A, Z) \to (A, Z+4) + 4e^{-},$$
 (1)

where lepton number conservation is violated by 4 units rather than 2 units. Though  $4\nu4\beta$  decay is allowed, the *Q*-dependent phase space factor ( $Q^{23}$ ) for eight outgoing particles severely impacts the decay rate, compared to  $Q^{11}$  for  $0\nu4\beta$  decay. The daughter (Z + 4) nucleus must of course have a smaller mass than the parent nucleus, so not all  $0\nu\beta\beta$  decay nuclei are good candidates. In fact, only three  $0\nu4\beta$  decay candidates have been identified:  ${}^{96}$ Zr,  ${}^{136}$ Xe, and  ${}^{150}$ Nd [1]. The *Q* value of  $0\nu4\beta$  decay of  ${}^{150}$ Nd to  ${}^{150}$ Gd is

The *Q* value of  $0\nu4\beta$  decay of <sup>150</sup>Nd to <sup>150</sup>Gd is 2.079 MeV, and is the largest of the three candidates. Competing decays include the two-neutrino double- $\beta$  ( $2\nu\beta\beta$ ) decay of <sup>150</sup>Nd ( $T_{1/2} = 8.2 \times 10^{18}$  years) and  $2\nu\beta\beta$  decay to excited

states ( $T_{1/2} = 1.2 \times 10^{20}$  years) (see Ref. [2] and references therein). Similarly, there is a possibility of the  $0\nu 4\beta$  decay proceeding via a  $0_1^+$  excited state of <sup>150</sup>Gd, which reduces the Q value by 1207.135 keV [3].

Though [1] estimates a half-life to be inordinately large, they emphasize that constraining this value experimentally is extremely valuable to provide inputs for new physics models. To this end, the NEMO-3 Collaboration has established a half-life limit for this decay to the ground state of <sup>150</sup>Gd [4]. We propose to search for the  $0\nu4\beta$  decay of <sup>150</sup>Nd to the  $0_1^+$  excited state of <sup>150</sup>Gd.

The daughter nucleus of the  $0\nu4\beta$  decay of <sup>150</sup>Nd is <sup>150</sup>Gd, which has a level scheme shown in Fig. 1.  $\beta$  decay to the 3<sup>-</sup> state is highly unlikely due to spin-parity considerations, so we will look for decays to the  $0_1^+$  state at 1207.135 keV. Decays to this state would result in a  $0_1^+ \rightarrow 2_1^+ \rightarrow 0_{g.s.}^+$  decay, emitting 569.031 and 638.050 keV  $\gamma$  rays in coincidence [3]. The rationale for concentrating on the  $0\nu4\beta$  decay to excited states is based on the fact that coincidence experiments, although less efficient, may provide greater sensitivity in searches for extremely rare events than singles measurements due to the reduction of background events.

## **II. EXPERIMENTAL METHOD**

Our experimental setup is optimized to search for coincident  $\gamma$  rays emitted from a disk-shaped source. Two

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FIG. 1. Level scheme of <sup>150</sup>Nd  $0\nu 4\beta$  decay to <sup>150</sup>Gd.

high-purity germanium (HPGe) detectors surrounded by several layers of active and passive shielding sandwich the source to be studied. These detectors are operated in both "singles" mode and coincidence mode, in order to detect  $\gamma$  rays emitted in subsequent decays of an excited state of a nucleus which has undergone a specific decay. The coincidence requirement of two  $\gamma$  rays of specific energies deposited simultaneously results in a significant reduction of random background events in the region of interest. Additionally, the uniqueness of the required coincident energies means there are few or no background candidates. These detectors are p-type coaxial germanium detectors with a diameter of 8.8 cm and thickness of 5.0 cm. They have nickel-plated magnesium endcaps that are 2.54 mm thick, and are connected to their cryostats in the j-type configuration. This configuration is ideal because it allows for the easy insertion of the crystals into the veto annulus. The preamplifier and high-voltage filter are located near the cryostat, away from the crystals, in order to reduce backgrounds.

Surrounding the HPGe detectors and sample is a sodium iodide (NaI) annulus with two plastic-plate scintillators on the endcaps. Three photomutiplier tubes (PMTs) on each end of the annulus and two PMTs on each plastic plate scintillator are operated in anticoincidence with the HPGe detectors to form an active veto. Next is a layered passive shield composed of 3/4 inch thick oxygen-free high conductivity (OFHC) copper plates followed by 6 inches of lead. For a full discussion of the electronics and data acquisition please see [5]. Finally, this setup was operated underground at the Kimballton Underground Research Facility (KURF) (details on KURF are in [6]). A drawing of the apparatus is shown in Fig. 2.

This setup was used to determine the half-life of  $2\nu\beta\beta$  decay of <sup>100</sup>Mo and <sup>150</sup>Nd to final excited states [5,7], and to determine a half-life limit of  $0\nu$ ECEC on <sup>112</sup>Sn [8]. More recently, the apparatus was used in searches for the  $2\nu\beta\beta$  decay of <sup>92</sup>Zr to excited states in <sup>96</sup>Mo [9] and for the single  $\beta$  decay of <sup>96</sup>Zr [10].

Here, we use data acquired from October 2008 to August 2010 for determination of  $2\nu\beta\beta$  decay of <sup>150</sup>Nd to final excited states [5]. The <sup>150</sup>Nd sample used in this experiment consists of 50.00 g of Nd<sub>2</sub>O<sub>3</sub> powder (43.09 ± 0.02 g Nd) enriched to 93.60% <sup>150</sup>Nd (40.33 ± 0.02 g <sup>150</sup>Nd). The mass of the <sup>150</sup>Nd

content is determined via isotopic analysis. This powder is compressed into a cylindrical cavity  $0.780 \pm 0.005$  cm thick and 2.86 cm in radius. This diameter sample makes the most of the coincidence efficiency of the detectors, which is higher near the center of the detectors. Data acquisition took place in 3- to 5-day-long runs over a period of 642.8 days.

# **III. COINCIDENCE EFFICIENCY**

The coincidence efficiency for this setup was determined in [7] using the decay of  $^{102}$ Ru, which emits two  $\gamma$  rays in coincidence in a  $0^+ \rightarrow 2^+ \rightarrow 0^+$  decay sequence. The energies of the  $\gamma$  rays in this decay are 468.64 and 475.10 keV. To adapt this efficiency measurement to our purposes, corrections need to be made to account for attenuation differences and the energy dependence of the efficiency. Full details on the corrections made to the efficiency for  $2\nu\beta\beta$  decay of  $^{150}$ Nd are given in [5]. Here, the corrections were recalculated for the energies of the  $0\nu4\beta$  decay of  $^{150}$ Nd. The final results yields a total coincidence efficiency for the  $0\nu4\beta$  decay of  $^{150}$ Nd of  $\epsilon_{\gamma\gamma} = (0.634 \pm 0.043)\%$ .



FIG. 2. Setup diagram for the double- $\beta$  decay apparatus. Please see text for details.



FIG. 3. The potential background peak at 569.70 keV is shown in a single-detector spectrum. Also note the lack of any excess of counts at 640 keV. See text for details of the origin of the background radiation.

## **IV. BACKGROUND CANDIDATES**

An extremely thorough discussion of potential background sources can be found in [5]. Here, we discuss the only option that contributes in the coincidence region of interest (ROI). As discussed in [5], there is a known contamination of  $^{232}$ Th decay products in the enriched <sup>150</sup>Nd sample. A search of the NNDC database [3] for coincident  $\gamma$  rays in the ROI reveals only one potential source that could reasonably be present in our setup. This transition is located in the  $\beta$  decay of <sup>228</sup>Ac to <sup>228</sup>Th. In this transition,  $\gamma$  rays of energy 570.91 keV (intensity  $I_{\nu} = 0.182\%$ ), 572.14 keV ( $I_{\nu} = 0.150\%$ ), and 640.32 keV ( $I_{\nu} = 0.054\%$ ) can be emitted. Indeed, in the single-detector spectrum, a peak can be observed at 569 keV as shown in Fig. 3, though no peak is obvious at 640 keV. To determine if background from this source is a concern, the level diagram of <sup>228</sup>Th must be inspected. The 640.32 keV  $\gamma$ ray originates from the 1901.92 keV energy level  $(J^{\pi} = 6^+)$ with a relative intensity of 48%. This transition proceeds through the 1261.57 keV (4<sup>+</sup>) level, and 36% of these transitions continue through the 1091.048 keV  $(4^+)$  level. From here, 2.3% of these decays result in a 571.8 keV  $\gamma$  ray. Therefore, it is unlikely there would be significant contamination from this decay in the coincidence spectrum. To check this, we analyzed 1.10 days of a ThO<sub>2</sub> sample inserted in the apparatus. As shown in Fig. 4, no events above background are shown in the coincidence ROI.

However, as mentioned above, there is a sizable peak around 569 keV. Investigation of the coincidence spectrum reveals that this peak occurs in coincidence with 1063 keV. This level transition occurs in the <sup>207</sup>Pb( $n, n'\gamma$ ) interaction when the excited <sup>207</sup>Pb nucleus decays to its ground state via the 1633.36 keV state, emitting 1063.66 and 569.70 keV  $\gamma$  rays in coincidence. However, this interaction requires a neutron flux, and is only apparent when the <sup>150</sup>Nd sample is present. We believe ( $\alpha, n$ ) reactions are taking place in the carbon in the teflon holder surrounding the <sup>150</sup>Nd sample. The  $\alpha$  decays are due to the <sup>232</sup>Th contamination in the <sup>150</sup>Nd sample itself. Though this peak is in our single- $\gamma$  ROI, no coincidence candidate exists.





FIG. 4. (a) The two-dimensional spectrum shows the ThO<sub>2</sub> data where no events are in coincidence above background in our region of interest (indicated by the dashed lines at 569.05 and 638.05 keV). (b) The singles spectrum for the ThO<sub>2</sub> data shows no peak at 569 keV. See text for details.

### V. ANALYSIS

The coincidence data is investigated by plotting events in detector 2 versus the events in detector 1. Only the events which meet the coincidence timing requirement (about 4  $\mu$ s) between the two detectors and the anticoincidence timing requirement (about 10  $\mu$ s) between the detectors and the veto are plotted in this spectrum. In the two-dimensional spectrum, the events of interest would occur at coordinates of (569.03 keV, 638.05 keV) and (638.05 keV, 569.03 keV). The significance of the counts in the two-dimensional regions of interest can best be seen by projecting them onto the x or y axis. To accomplish this, an energy condition (i.e.,  $569.03 \pm 2.50$  keV) is applied to events in one detector, and all the events which occur in the other detector in coincidence are projected into a histogram. This results in four histograms; two are in coincidence with 569.03 keV in detectors 1 and 2, and two are in coincidence with 638.05 keV in detectors 1 and 2. The corresponding histograms can then be summed so that



FIG. 5. Coincidence data for decays to the  $0_1^+$  state in <sup>150</sup>Gd. Spectrum (a) is in coincidence with 569.03 keV, and spectrum (b) is in coincidence with 638.05 keV. The coincidence-energy window in spectra (a) and (b) is 2.50 keV for detector 2 and 3.00 keV for detector 1. Events within the region of interest are colored red.

all events in coincidence with a 569.03 keV photon are in one histogram, and vice versa, reducing the number of histograms to two (see Fig. 5). To avoid any histogram summing effects, the detectors were carefully gain matched and the bin widths are extremely close.

## VI. QUADRUPLE- $\beta$ DECAY OF <sup>150</sup>Nd TO THE 0<sup>+</sup><sub>1</sub> EXCITED STATE OF <sup>150</sup>Gd

Over 642.8 days of data acquisition, seven events in the 569–638 keV coincidence window are observed with an average background of  $5.48 \pm 0.57$  counts (see Fig. 5), therefore, a half-life limit can be set. The half-life limit is given by

$$T_{1/2} > \frac{\ln(2)N_0 t f_b \epsilon_{\gamma\gamma}^{\text{tot}}}{N_d},\tag{2}$$

Here,  $N_0$  is the number of <sup>150</sup>Nd nuclei in the sample, *t* is total counting time (642.8 days or 1.76 years),  $\epsilon_{\gamma\gamma}^{\text{tot}}$  is the total coincidence detection efficiency for the 569.08–638.05 keV cascade, and  $f_b$  is the branching ratio for the particular cascade. The value of  $N_0$  is  $1.620 \pm 0.003 \times 10^{23}$  nuclei of <sup>150</sup>Nd. The coincidence efficiency for the 569.08–638.05 keV cascade is  $\epsilon_{\gamma\gamma} = (0.634 \pm 0.043)\%$ , and  $f_b$  is 100% for this cascade.  $N_d$  is a value chosen using the TRolke class in the ROOT analysis framework [11]. This class considers a Poisson signal in the presence of an uncertain background and efficiency, and computes the confidence interval for this situation, using the method outlined in [12]. For the calculation described here, we used a background estimate simultaneously measured from the sidebands of our signal, and assumed a Poisson uncertainty in this estimate, with a Gaussian uncertainty in the efficiency measurement. Here, with an observed signal of 7 events with a background of  $5.48 \pm 0.57$  events in the region of interest, the Rolke method gives an upper limit to the profile likelihood of  $N_d/\epsilon_{\gamma\gamma}^{tot} = 1123$  events, resulting in a half-life limit of  $T_{1/2} > 1.76 \times 10^{20}$  years (90% CL). This limit was obtained with only 40.33 g of <sup>150</sup>Nd; significant improvements to the limit could be achieved with a larger sample.

# **VII. CONCLUSIONS**

We have presented the first search for  $0\nu 4\beta$  decay of <sup>150</sup>Nd to excited final states of <sup>150</sup>Gd. Though this decay is predicted to have a half-life many orders of magnitude larger than this limit, it is important to experimentally constrain these quantities to check for potential enhancements to the decay rate, as discussed in [1].

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