Double-Beta Decay of $^{96}\text{Zr}$ and Double-Electron Capture of $^{156}\text{Dy}$ to Excited Final States

by

Sean W. Finch

Department of Physics
Duke University

Date: ______________________

Approved:

__________________________
Werner Tornow, Supervisor

__________________________
Calvin Howell

__________________________
Kate Scholberg

__________________________
Berndt Mueller

__________________________
Albert Chang

Dissertation submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy in the Department of Physics in the Graduate School of Duke University 2015
Abstract

Double-Beta Decay of $^{96}$Zr and Double-Electron Capture of $^{156}$Dy to Excited Final States

by

Sean W. Finch

Department of Physics
Duke University

Date: ______________________

Approved:

________________________
Werner Tornow, Supervisor

________________________
Calvin Howell

________________________
Kate Scholberg

________________________
Berndt Mueller

________________________
Albert Chang

An abstract of a dissertation submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy in the Department of Physics in the Graduate School of Duke University

2015
Abstract

Two separate experimental searches for second-order weak nuclear decays to excited final states were conducted. Both experiments were carried out at the Kimballton Underground Research Facility to provide shielding from cosmic rays. The first search is for the $2\nu\beta\beta$ decay of $^{96}\text{Zr}$ to excited final states of the daughter nucleus, $^{96}\text{Mo}$. As a byproduct of this experiment, the $\beta$ decay of $^{96}\text{Zr}$ was also investigated. Two coaxial high-purity germanium detectors were used in coincidence to detect $\gamma$ rays produced by the daughter nucleus as it de-excited to the ground state. After collecting 1.92 years of data with 17.91 g of enriched $^{96}\text{Zr}$, half-life limits at the level of $10^{20}$ yr were produced. Measurements of this decay are important to test $0\nu\beta\beta$-decay nuclear matrix element calculations, which are necessary to extract the neutrino mass from a measurement of the $0\nu\beta\beta$ decay half-life.

The second experiment is a search for the resonantly-enhanced neutrinoless double-electron capture decay of $^{156}\text{Dy}$ to excited states in $^{156}\text{Gd}$. Double-electron capture is a possible experimental alternative to $0\nu\beta\beta$ decay, which could distinguish the Dirac or Majorana nature of the neutrino. Two clover high-purity germanium detectors were used in coincidence to investigate the decay. A 213.5 mg enriched $^{156}\text{Dy}$ sample was observed for 0.635 year, producing half-life limits of $10^{17}$ yr. The limits produced by both of these experiments are currently the most stringent limits available for these decays.
Contents

Abstract iv
List of Tables ix
List of Figures xi
List of Abbreviations and Symbols xiv
Acknowledgements xvi

1 Introduction 1
  1.1 Neutrinos and the Standard Model 2
      1.1.1 Majorana and Dirac neutrinos 4
  1.2 Weak nuclear decays 7
  1.3 $\beta\beta$ decay 9
      1.3.1 Excited-state decays 13
  1.4 Double-electron capture 15
  1.5 Experimental technique 18

2 Theory 20
  2.1 $\beta$-decay theory 20
      2.1.1 Decay-rate calculation 21
  2.2 $\beta\beta$ decay theory 25
      2.2.1 $2\nu\beta\beta$ decay-rate calculation 26
      2.2.2 $0\nu\beta\beta$ decay-rate calculation 28
2.2.3 $\beta\beta$ Nuclear Matrix Elements ........................................ 32

2.3 ECEC theory ................................................................. 39
  2.3.1 $EC$ decay-rate calculation ............................................ 39
  2.3.2 $ECEC$ decay-rate calculation ........................................ 40
  2.3.3 $0\nu ECEC$ NME calculations ......................................... 43

3 Experimental Technique ....................................................... 45
  3.1 $\gamma$-ray detection ...................................................... 45
    3.1.1 HPGe detectors ...................................................... 47
    3.1.2 NaI annulus ......................................................... 50
  3.2 Two-coaxial HPGe apparatus ............................................ 52
    3.2.1 Electronics .......................................................... 53
  3.3 Two-clover HPGe apparatus ............................................. 54
    3.3.1 Clover HPGe detectors .............................................. 55
    3.3.2 Active shielding .................................................... 56
    3.3.3 Passive shielding .................................................. 58
    3.3.4 Electronics ........................................................ 59
  3.4 KURF ................................................................. 62
  3.5 Samples ................................................................. 62
    3.5.1 $^{96}$Zr sample ...................................................... 64
    3.5.2 $^{156}$Dy sample ...................................................... 66
  3.6 Previous measurements ................................................... 66
    3.6.1 On $^{96}$Zr ............................................................... 67
    3.6.2 On $^{156}$Dy ............................................................. 68

4 $^{96}$Zr analysis ................................................................. 70
  4.1 Data collection and quality control .................................... 70
4.2 Data processing ........................................ 73
4.3 $\beta\beta$-decay results ................................ 77
4.4 $\beta\beta$-decay efficiency calculation .............. 82
  4.4.1 Energy dependence ................................ 83
  4.4.2 Target attenuation ................................. 84
  4.4.3 Detector separation ................................. 86
  4.4.4 Z dependence of the efficiency ................. 87
  4.4.5 Angular correlation of coincident $\gamma$s ...... 87
  4.4.6 Final efficiency .................................. 88
4.5 $\beta$-decay results .................................. 90
4.6 $\beta$-decay efficiency calculation ................. 94

5 $^{156}$Dy analysis ........................................ 99
  5.1 Data collection ...................................... 99
  5.2 Data processing ..................................... 100
  5.3 Data analysis ....................................... 102
    5.3.1 To the 1946.4 keV state ....................... 103
    5.3.2 To the 1952.4 keV state ....................... 104
    5.3.3 To the 1988.5 keV state ....................... 106
    5.3.4 To the 2003.7 keV state ....................... 106
  5.4 Efficiency measurements ......................... 108
    5.4.1 Energy dependence ............................ 112
    5.4.2 Addback factor ................................ 114
    5.4.3 Target attenuation ............................ 116
    5.4.4 Detector separation and source geometry .... 116
    5.4.5 Angular dependence of coincident $\gamma$ rays .. 117
5.4.6 Final efficiency ........................................ 118
5.4.7 Contributions from three-γ decays .................. 118

6 Conclusion .................................................. 121

6.1 Limit setting .............................................. 121
6.2 ββ decays of $^{96}$Zr to excited final states ............. 122
6.3 β decay of $^{96}$Zr ........................................ 125
6.4 $ECEC$ decays of $^{156}$Dy ................................ 127
6.5 Concluding remarks ..................................... 130

Bibliography .................................................. 132

Biography ..................................................... 137
List of Tables

1.1 $\beta\beta$-decay phase-space factors ........................................... 12
1.2 $\beta\beta$-decay experimental half-lifes .......................................... 13
1.3 $\beta\beta$ decay to $0^+_1$ phase space ........................................... 15
2.1 $\beta$-decay transitions ............................................................... 24
2.2 Resonant-$EC\!\!\!\!\!\!\!\!E\!\!C$ decays in $^{156}$Dy ................................ 43
2.3 $0\nu E\!\!\!\!\!\!\!\!E\!\!C\!\!\!\!\!\!\!C$ NMEs for $^{156}$Dy ............. 44
3.1 Isotopic enrichment for ZrO$_2$ samples ....................................... 65
3.2 Isotopic enrichment for Dy$_2$O$_3$ samples .................................... 66
3.3 Limits on $\beta\beta$ decay of $^{96}$Zr to excited states at CL = 90% ....... 68
3.4 Limits on $E\!\!C\!\!\!\!\!\!\!E\!\!C$ decay of $^{156}$Dy to excited states at CL = 90% .................. 69
4.1 Efficiency and background in the region of interest ......................... 74
4.2 Count rate for the $^{96}$Zr ROI during background and $^{150}$Nd runs ....... 82
4.3 Efficiency correction ratios for different coincidences in $^{96}$Mo ........ 86
4.4 Systematic error budget for $\beta\beta$ decay of $^{96}$Zr ....................... 90
4.5 Three most probably decay sequences for the $5^+$ state of $^{96}$Mo ...... 91
4.6 Results of search for $^{96}$Zr’s $\beta$ decay .................................... 94
4.7 Systematic error budget for the $\beta$ decay of $^{96}$Zr ....................... 98
5.1 Coincident efficiency of the two-clover apparatus as measured with $^{102}$Rh ................................................................. 112
5.2 Efficiency correction ratios for different coincidences in $^{156}$Dy ........ 117
<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.3 Efficiency of the two-clover apparatus for different angular distribution</td>
<td>118</td>
</tr>
<tr>
<td>5.4 Corrected efficiency for the $^{156}$Dy ROIs.</td>
<td>118</td>
</tr>
<tr>
<td>5.5 Systematic error budget for the $ECEC$ decay of $^{156}$Dy</td>
<td>119</td>
</tr>
<tr>
<td>5.6 Contributions to the ROI from the three-$\gamma$ decays</td>
<td>120</td>
</tr>
<tr>
<td>6.1 Sensitivity of $\beta\beta$ decay to excited states in $^{96}$Zr</td>
<td>123</td>
</tr>
<tr>
<td>6.2 Half-life limits on the $\beta\beta$ decay of $^{96}$Zr to excited states</td>
<td>124</td>
</tr>
<tr>
<td>6.3 Half-life limits for $^{96}$Zr’s $\beta$ decay</td>
<td>126</td>
</tr>
<tr>
<td>6.4 Combined statistical limits for $^{96}$Zr’s $\beta$ decay</td>
<td>127</td>
</tr>
<tr>
<td>6.5 Statistical sensitivity for the $^{156}$Dy $ECEC$ ROIs.</td>
<td>128</td>
</tr>
<tr>
<td>6.6 Final half-life limits for the $ECEC$ of $^{156}$Dy to excited states</td>
<td>129</td>
</tr>
<tr>
<td>6.7 Combined statistical limits for the $ECEC$ decay of $^{156}$Dy to the 2003.7 keV state.</td>
<td>129</td>
</tr>
</tbody>
</table>
# List of Figures

1.1 The four states of the Dirac neutrino ........................................ 5
1.2 The two states of the Majorana neutrino ................................. 5
1.3 The $A = 156$ isobar ............................................................. 9
1.4 $2\nu\beta\beta$ decay ............................................................... 9
1.5 $0\nu\beta\beta$ decay ............................................................... 11
1.6 $^{96}\text{Zr }\beta\beta$ decay to excited states .............................. 14
1.7 Resonant $0\nu ECEC$ in $^{156}\text{Dy}$ to an excited state in $^{156}\text{Gd.}$ ....... 16
1.8 $0\nu ECEC$ decay mediated by exchange of a virtual neutrino ......... 17
1.9 Coincidence detection principle .............................................. 19
2.1 The single-$\beta$ decay of $^{96}\text{Zr}$ ........................................... 25
2.2 NME for $0\nu\beta\beta$ ............................................................. 34
2.3 NME for $2\nu\beta\beta$ ............................................................. 38
2.4 Ratio of ground-state to excited-state NME ............................... 39
3.1 Spectra for coaxial HPGe ....................................................... 48
3.2 Coaxial-HPGe schematic ....................................................... 48
3.3 Two-coaxial HPGe apparatus ............................................... 53
3.4 Two-clover HPGe apparatus .................................................. 56
3.5 Clover segments ................................................................. 57
3.6 NaI Annulus ................................................................. 58
3.7 Lead shielding ................................................................. 59
<table>
<thead>
<tr>
<th>Section</th>
<th>Title</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.8</td>
<td>Clover TDC and trigger circuit</td>
<td>60</td>
</tr>
<tr>
<td>3.9</td>
<td>Clover ADC circuit</td>
<td>61</td>
</tr>
<tr>
<td>3.10</td>
<td>Clover NaI circuit</td>
<td>61</td>
</tr>
<tr>
<td>3.11</td>
<td>Muon flux at KURF</td>
<td>63</td>
</tr>
<tr>
<td>3.12</td>
<td>Spectra from KURF</td>
<td>63</td>
</tr>
<tr>
<td>3.13</td>
<td>KURF lab</td>
<td>64</td>
</tr>
<tr>
<td>3.14</td>
<td>Two-clover housing</td>
<td>65</td>
</tr>
<tr>
<td>4.1</td>
<td>$^{96}$Zr detector live time</td>
<td>71</td>
</tr>
<tr>
<td>4.2</td>
<td>FWHM of coaxial HPGe detectors over time</td>
<td>71</td>
</tr>
<tr>
<td>4.3</td>
<td>$^{40}$K yield as a function of time</td>
<td>72</td>
</tr>
<tr>
<td>4.4</td>
<td>$^{214}$Bi yield as a function of time</td>
<td>73</td>
</tr>
<tr>
<td>4.5</td>
<td>Long term detector stability</td>
<td>74</td>
</tr>
<tr>
<td>4.6</td>
<td>Two-coaxial HPGe veto TAC spectra</td>
<td>75</td>
</tr>
<tr>
<td>4.7</td>
<td>Two-coaxial HPGe TAC coincidence spectra</td>
<td>76</td>
</tr>
<tr>
<td>4.8</td>
<td>Two-coaxial HPGe 2D coincidence spectra</td>
<td>76</td>
</tr>
<tr>
<td>4.9</td>
<td>Detector resolution during $^{96}$Zr runs</td>
<td>77</td>
</tr>
<tr>
<td>4.10</td>
<td>Background estimation for the decay to the $0^+_1$ state</td>
<td>78</td>
</tr>
<tr>
<td>4.11</td>
<td>The ROI for the $\beta\beta$ decay of $^{96}$Zr to the $0^+_1$ state</td>
<td>79</td>
</tr>
<tr>
<td>4.12</td>
<td>$\beta\beta$ decay in $^{96}$Zr to different excited states of $^{98}$Mo</td>
<td>80</td>
</tr>
<tr>
<td>4.13</td>
<td>The ROI for the $\beta\beta$ decay of $^{96}$Zr to higher excited states</td>
<td>81</td>
</tr>
<tr>
<td>4.14</td>
<td>Decay scheme for $^{102}$Rh</td>
<td>83</td>
</tr>
<tr>
<td>4.15</td>
<td>Energy dependent efficiency for coaxial HPGe detectors</td>
<td>84</td>
</tr>
<tr>
<td>4.16</td>
<td>GEANT4 coaxial HPGe</td>
<td>84</td>
</tr>
<tr>
<td>4.17</td>
<td>GEANT4 $^{96}$Zr sample</td>
<td>85</td>
</tr>
<tr>
<td>4.18</td>
<td>GEANT4 two-coaxial apparatus</td>
<td>85</td>
</tr>
</tbody>
</table>
4.19 Angular correlation distributions .............................. 88
4.20 Radial efficiency for the two-coaxial apparatus ............... 89
4.21 $^{96}$Zr $\beta$ decay ROI histograms .............................. 93
4.22 GEANT4 two-coaxial HPGe apparatus with NaI ................ 95
4.23 Position-dependent efficiency for $^{96}$Zr’s $\beta$ decay ............... 97
5.1 Clover resolution during $^{156}$Dy runs .......................... 101
5.2 Clover spectra during $^{156}$Dy runs .............................. 101
5.3 Decay scheme for the 1946.4 keV state of $^{156}$Gd ............... 103
5.4 ROI for the 1857.4+89.97 keV ECEC coincidence ............... 104
5.5 Decay scheme for the 1952.4 keV state of $^{156}$Gd ............... 105
5.6 ROI for the 1242.5+709.9 keV ECEC coincidence ............... 105
5.7 ROI for the 1899.5+89.97 keV ECEC coincidence ............... 107
5.8 Decay scheme for the 2003.7 keV state of $^{156}$Gd ............... 108
5.9 ROI for ECEC to the 2003.7 keV state ....................... 109
5.10 Average coincident efficiency of the two-clover apparatus ....... 111
5.11 Coincident efficiency of the two-clover apparatus along the $x$-axis .. 111
5.12 Coincidence efficiency of the two-clover apparatus as a function of position with the omission of c13 .................. 112
5.13 GEANT4 model of clover detector ............................... 113
5.14 Energy dependent efficiency for coaxial HPGe detectors ........... 113
5.15 Addback ratio for the clover detectors ....................... 115
5.16 Addback ratio position dependence ............................ 116
List of Abbreviations and Symbols

Symbols

\[ {}^A_ZX_N \] Nucleus with \( Z \) protons, \( N \) neutrons, and atomic number \( A = Z + N \)

\( \beta \) Single-\( \beta \) decay

\( 2\nu\beta\beta \) Two-neutrino double-\( \beta \) decay

\( 0\nu\beta\beta \) Neutrinoless double-\( \beta \) decay

\( EC \) Electron capture

\( ECEC \) Double-electron capture

\( 0\nuECEC \) Neutrinoless double-electron capture

Abbreviations

ADC Analog-to-Digital Converter

CFD Constant Fraction Discriminator

DAQ Data Acquisition [system]

ECL Emitter-Coupled Logic

EF Enhancement Factor (for resonant-\( ECEC \))

FWHM Full Width at Half Maximum

GEANT4 GEometry ANd Tracking 4, a Monte Carlo simulation software

HPGe High-purity Germanium [detector]

IBM Interacting Boson Model

ISM Interacting Shell Model
<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>KURF</td>
<td>Kimballton Underground Research Facility</td>
</tr>
<tr>
<td>LNGS</td>
<td>Gran Sasso National Laboratory</td>
</tr>
<tr>
<td>NaI</td>
<td>Sodium Iodide</td>
</tr>
<tr>
<td>NIM</td>
<td>Nuclear Instrument Module</td>
</tr>
<tr>
<td>NME</td>
<td>Nuclear Matrix Element</td>
</tr>
<tr>
<td>ORNL</td>
<td>Oak Ridge National Laboratory</td>
</tr>
<tr>
<td>PMT</td>
<td>Photomultiplier Tube</td>
</tr>
<tr>
<td>QRPA</td>
<td>Quasiparticle Random Phase Approximation</td>
</tr>
<tr>
<td>ROI</td>
<td>Region Of Interest</td>
</tr>
<tr>
<td>TAC</td>
<td>Time-to-Analog Converter</td>
</tr>
<tr>
<td>TDC</td>
<td>Time-to-Digital Converter</td>
</tr>
<tr>
<td>TFA</td>
<td>Timing-Filtering Amplifier</td>
</tr>
<tr>
<td>TUNL</td>
<td>Triangle Universities Nuclear Laboratories</td>
</tr>
<tr>
<td>VME</td>
<td>VERSA module Eurocard</td>
</tr>
</tbody>
</table>
Acknowledgements

“It takes a village to raise a child. It takes a good advisor, numerous professors, half a dozen technical staff, a few good post-docs, a support group of fellow graduate students, four universities, and lots of DOE funding to raise a PhD student.”

- African Proverb

I owe the most thanks to my advisor, Werner Tornow, for facilitating this research and my growth as a physicist. I would like to thank Cavlin Howell, whose work as director of TUNL has helped create an excellent environment for graduate students to learn and develop the wide range of skills necessary for a complete education in experimental nuclear physics. I would also like to thank Bruce Vogelaar and Derek Rountree of Virginia Tech for their work in organizing and maintaining the Kimballton Underground Research Facility, where this research was conducted. Rajarshi Raut helped with the initial construction of the two-clover apparatus and taught me so much during my first two years of graduate school. I am thankful for the TUNL technical staff: Bret Carlin, Richard O’Quinn, John Dunham, Chris Westerfeldt, and Brian Walsh. This work would not be possible without their assistance, which is too long to list here. On a personal note, I would like to thank grads09 who made graduate school fun and provided lots of helpful advice. Last, but certainly not least, to my parents, Charles and Jody, and my soon-to-be wife, Heather, for all they have done over the years.

xvi
This thesis investigates two separate rare decays: the double-$\beta$ decay of $^{96}$Zr and the double-electron capture of $^{156}$Dy to excited final nuclear states. The interest in these second-order weak nuclear decays is due to their implications for neutrino physics. If realized in nature, they allow us to unambiguously distinguish the Majorana or Dirac nature of the neutrino and make a measurement of the neutrino mass. This chapter gives a brief history of neutrino physics, which fittingly began with observations of the first-order weak nuclear decays. This leads to the current theoretical description of neutrinos by the Standard Model, including an explanation of the differences between Dirac and Majorana neutrinos. A qualitative description of double-$\beta$ decay and double-electron capture is then given, with the quantitative theory detailed in Chapter 2. Lastly, current experimental efforts are summarized, including the experimental technique used in this work. The significance of the decays studied in this thesis is emphasized when appropriate.
1.1 Neutrinos and the Standard Model

The conception of weak decays and the neutrino began with the study of $\beta$ decay. $\beta$ decay was first theorized as a 2-body decay: a neutron decays into a proton with emission of an electron. Kinematics then dictate that the emitted electron must be monoenergetic, which conflicts with experimental observations of a continuous electron spectrum. In 1930 Pauli hypothesized the existence of a third particle in the decay channel [1]. If this hypothetical particle was electrically neutral and had a small or zero mass, it would solve the electron energy spectrum problem. After the discovery of the neutron, a spin-1/2 neutrino was necessary to solve the spin discrepancies of $\beta$ decay as well.

Fermi named the hypothetical particle the neutrino and included it in his 1934 theory of $\beta$ decay [2]. This was the first formulation of the weak interaction, although we now know it was only an effective theory. Using Fermi’s theory, Bethe and Peierls estimated a neutrino-matter cross section of $\sigma < 10^{-44}$ cm$^2$ and concluded that “there is no practically possible way of observing the neutrino” [3]. This claim stood for nineteen years until the first experimental observation of neutrinos. In 1953 Reines and Cowan [4] detected neutrinos from a nuclear reactor using the inverse $\beta$-decay reaction

$$\bar{\nu}_e + p \rightarrow n + e^+.$$ (1.1)

In 1957 a pair of experiments using weak nuclear decays provided new fundamental information about the weak interaction and the neutrino. First, parity violation was experimentally verified using the $\beta$ decay of $^{60}$Co [5]. Later, the electron-capture decay of $^{152m}$Eu was used to show that the neutrino is left-handed [6]. These discoveries were used to formulate the theory of weak interactions and the Standard Model of particle physics. In this model, the weak currents, mediated by the $W^\pm$ and $Z^0$ bosons, only couple left-handed states. This results in the weak interaction
only producing left-handed neutrinos or right-handed antineutrinos.

Danby et al. proved in 1962 that neutrinos produced in association with muons were distinct from those produced in association with an electron [7]. This led to the notion of neutrino flavors. Each neutrino flavor is a distinct particle, with the flavor defined by the lepton produced when the neutrino is created or interacts. The later discovery of the τ and its associated neutrino gives the three current neutrino flavors: the electron neutrino $\nu_e$, muon neutrino $\nu_\mu$, and tau neutrino $\nu_\tau$. The three lepton generations can be grouped with the three quark generations to produce the three generations of matter:

$$
\begin{array}{ccc}
  u & c & t \\
  d & s & b \\
  e & \mu & \tau \\
  \nu_e & \nu_\mu & \nu_\tau \\
\end{array}
$$

(1.2)

Neutrinos were assumed to be massless by the Standard Model, but recent observations of neutrino oscillations using atmospheric [8], solar [9], and reactor [10] neutrinos proved that neutrinos have mass. Neutrino oscillations were first theorized by Maki, Nakagawa, and Sakata in 1962 [11] and are only possible for massive neutrinos. The oscillation phenomenon arises if the neutrino’s flavor states $\nu_\alpha$ are a superposition of its mass states $\nu_i$

$$
\nu_\alpha = \sum_{i=0}^{N} U_{\alpha i} \nu_i ,
$$

(1.3)

where $U_{\alpha i}$ is the neutrino mixing matrix. Neutrinos are created in pure flavor states but propagate as a function of their mass states. These mass states interfere with each other as the neutrino travels. This allows neutrinos of one flavor to transform into a different flavor as they travel. The transformation can be shown to exhibit an oscillatory behavior, thus warranting the name neutrino oscillations. Experiments observing neutrino oscillations are highly sensitive to the mass squared differences
of three neutrino mass states \( \delta m^2_{ij} \equiv m^2_i - m^2_j \). These experiments, however, cannot measure the absolute neutrino mass.

Neutrinoless double-\( \beta \) decay allows for a measurement of the absolute neutrino mass, as will be discussed in later sections. Direct searches for neutrino mass are also done by investigating the \( \beta \) decay of tritium and searching for changes in the endpoint of the electron spectrum. These experiments currently have produced a lower limit of \( M_{\nu e} < 2.3 \text{ eV} \) [12]. This motivates the question: why is the neutrino mass six orders of magnitude smaller than the next lightest lepton?

1.1.1 Majorana and Dirac neutrinos

The Dirac equation produces two sets of solutions, now referred to as particles and antiparticles. For electrons and other charged particles, the distinction between these two states is obvious. The same cannot be said for an electrically neutral particle, such as the neutrino. The neutrino was assumed to be described by the Dirac equation and has a distinct antiparticle state, the antineutrino, in analog to the electron. In 1937 Majorana proposed a formal theory for fermions that are identical to their antiparticle [13]. These particles are now known as Majorana particles. There exist other particles who are their own antiparticle, such as the \( \pi^0 \) or \( K^0 \), but these particles are both bosons and not fundamental particles. Thus, the neutrino is the only Majorana particle candidate. Majorana particles must be invariant under charge conjugation. This results in the first distinction between Majorana and Dirac particles: Majorana particles must possess a zero electric and magnetic dipole moment. This is not the case for Dirac neutrinos, which may possess dipole moments.

The distinction between the Majorana and Dirac particles is illustrated by the following argument assuming Lorentz and CPT (charge-parity-time) invariance. Assume a massive left-handed Dirac neutrino \( \nu_L \), as shown in Fig. 1.1. When acted
on by the CPT operator, we produce a right-handed antineutrino $\bar{\nu}_R$. Assuming the neutrino is massive, there exists a frame of reference traveling faster than the neutrino (yet slower than the speed of light). By Lorentz boosting into this frame the neutrino’s direction will change with its spin state unchanged, flipping the neutrino’s handedness. This produces a right-handed neutrino $\nu_R$ and a left-handed antineutrino $\bar{\nu}_L$. These two states are CPT transforms of each other and will not interact in the Standard Model.

Now we assume a left-handed Majorana neutrino $\nu_L$, as shown in Fig. 1.2. Once again the Lorentz operator will produce a right-handed neutrino $\nu_R$. The CPT operator, when acted on $\nu_L$, will now produce a right-handed neutrino $\nu_R$. For Majorana neutrinos, only two distinct states exist, as compared to the four distinct states of the Dirac neutrino.

The above argument relies on neutrinos having a rest mass. If the neutrino were massless, the left and right-handed states would be distinct. Since the weak interaction only involves left-handed currents, there is no reason for $\nu_R$ and $\bar{\nu}_L$ to exist in the massless Dirac case, leaving only two neutrino states. As such, there is
no distinction between massless Majorana and Dirac neutrinos [14]. This is one of
the reasons that searches for double-β decay and Majorana neutrinos increased after
the discovery of neutrino oscillations and the proof of massive neutrinos.

A further study of Majorana and Dirac neutrinos is done by investigating their

\[ -L^{\text{Dirac}}_M = \bar{\nu}_L M_D \nu_R \]  

\[ = \frac{1}{2} (\bar{\nu}_L M_D \nu_R + \bar{\nu}_L^{\text{c}} M_D^T \nu_R^{\text{c}}) , \]  

where \( ^c \) denotes charge conjugation and \( M_D \) is the Dirac mass. In the full neutrino
Lagrangian, \( M_D \) is a matrix containing the three neutrino mass states and the wave
functions \( \nu \) are vectors of the three mass states. In the second line, this equation
is expanded into two identical terms for reasons that will be seen later. This La-
grangian, with the associated kinetic and weak interaction terms, is invariant under
the global phase transformation

\[ \nu(x) \to e^{i\Phi} \nu(x) ; \quad \nu(x)^c \to e^{-i\Phi} \nu(x)^c ; \]

\[ \bar{\nu}(x) \to e^{-i\Phi} \bar{\nu}(x) ; \quad \left( \bar{\nu}(x) \right)^c \to e^{i\Phi} \left( \bar{\nu}(x) \right)^c . \]  

This symmetry will have a corresponding conserved quantity, known as lepton num-
ber. Neutrinos are currently written into the Standard Model with Dirac mass terms
and as such, conservation of lepton number is built into the Standard Model.

The Majorana mass term is written as

\[ -2L^{\text{Majorana}}_M = \bar{\nu}_L M_L \nu_L^c + \bar{\nu}_R^{\text{c}} M_R \nu_R . \]  

This mass Lagrangian, however, is not invariant under the phase transformation
given by Eq. (1.6). As such, Majorana mass terms for the neutrino would violate
lepton number conservation and require changes to the Standard Model.
In general, the neutrino could be a linear superposition of a Dirac and Majorana particle, leading to the most general Lagrangian mass term

\[-2\mathcal{L}_M = \overline{\nu}_L M_D \nu_R + \overline{\nu}_L^c M_D^T \nu_R^c + \overline{\nu}_L M_L (\nu_L)^c + \overline{\nu}_R^c M_R \nu_R \]

\[= \left[ \begin{array}{c} \overline{\nu}_L \\ (\nu_R)^c \end{array} \right] \left( \begin{array}{cc} M_L & M_D \\ M_D^T & M_R \end{array} \right) \left[ \begin{array}{c} (\nu_L)^c \\ \nu_R \end{array} \right]. \tag{1.8} \]

This formulation is frequently used to explain the mass discrepancy between neutrinos and charged leptons. The physically observed neutrino masses $M_\nu$ would be found by diagonalizing the matrix in Eq. (1.8). It is usually assumed that $M_D$ is characterized by the electroweak symmetry breaking scale, or the vacuum expectation value of the Higgs $\langle H \rangle$. This gives $M_D \approx \langle H \rangle \approx 125 \text{ GeV}$, with other reasonable assumptions giving $M_L \ll \langle H \rangle$, and $M_R \gg \langle H \rangle$ [15]. Using these values, we can calculate the observed neutrino masses

\[M_\nu \approx M_L - M_D M_R^{-1} M_D^T . \tag{1.9} \]

This is known as the seesaw relationship. In the type-I seesaw model, $M_L = 0$, and larger values of $M_R$ can force the observed neutrino mass $M_\nu$ to be small yet nonvanishing. Many other seesaw models and mechanisms have been proposed, but they mostly require Majorana neutrino mass terms and lepton number violation.

1.2 Weak nuclear decays

The weak nuclear decays, $\beta^+$, $\beta^-$, and $EC$, are the most common form of radioactive disintegration. In these decays, the nucleus moves across an isobar, changing the nuclear charge while preserving the total number of nucleons. The common $\beta^-$, $\beta^+$, and $EC$ decays may be represented by

\[^A_Z X_N \rightarrow ^A_{Z+1} Y_{N-1} + e^- + \bar{\nu}_e , \tag{1.10} \]

\[^A_Z X_N \rightarrow ^A_{Z-1} Y_{N+1} + e^+ + \nu_e , \tag{1.11} \]

\[e^- + ^A_Z X_N \rightarrow ^A_{Z-1} Y_{N+1} + \nu_e , \tag{1.12} \]
respectively, where $Z$ is the number of protons, $N$ the number of neutrons, and the mass $A = Z + N$. The $Q$ value of this reaction is the difference in mass of the parent and daughter nuclei. For $\beta^-$ decay this is

$$Q_\beta = M_{Z,A} - M_{Z+1,A}. \quad (1.13)$$

In order to investigate the $Q$ value of $\beta$ decay, we turn to the liquid drop model which approximates the binding energy of different nuclei. The model is as follows:

$$B(Z, A) = a_V A - a_S A^{2/3} - a_C Z^2 A^{-1/3}$$

$$- a_A \left( \frac{Z - A/2}{A} \right)^2 + a_P \frac{(-1)^Z + (-1)^N}{2} A^{-1/2}, \quad (1.14)$$

where $a_n$ are constants determined from an empirical fit to data. The equation includes a volume term $a_V$, a surface area term $a_S$, a Coulomb term $a_C$, a term to discourage neutron-proton asymmetry $a_A$, and a pairing term for unpaired protons or neutrons $a_P$. This equation is quadratic in $Z$, resulting in a parabola. For odd $A$ this parabola will have one minimum. Nuclei to the left will $\beta^-$ decay while nuclei to the right will $\beta^+$ or $EC$ to reach the minimum. For even $A$, the pairing term creates two parabolas, one for even-even nuclei and one for odd-odd nuclei. This occasionally results in local minima where a first order process, such as $\beta$ decay, is not energetically allowed but a second order process, $\beta\beta$ decay, is allowed. This can be seen in Fig. 1.3, which shows the $A = 156$ isobar. Note that $^{156}$Dy is energetically forbidden to decay via $EC$, but could decay to $^{156}$Gd if a second order process changed the nuclear charge by two units. Analogous to Eq. (1.10), we write the
second order processes $\beta^+\beta^+$, $\beta^+EC$, and $ECEC$ as

\begin{align}
\frac{A}{2}X_N &\rightarrow \frac{A}{Z+2}Y_{N-2} + 2e^- + 2\bar{\nu}_e, \quad (1.15) \\
\frac{A}{2}X_N &\rightarrow \frac{A}{Z-2}Y_{N+2} + 2e^- + 2\nu_e, \quad (1.16) \\
e^- + \frac{A}{2}X_N &\rightarrow \frac{A}{Z-2}Y_{N+2} + e^+ + 2\nu_e, \quad (1.17) \\
2e^- + \frac{A}{2}X_N &\rightarrow \frac{A}{Z-2}Y_{N+2} + 2\nu_e, \quad (1.18)
\end{align}

respectively. Subsequent analysis of the $\beta$-decay $Q$ values produces 35 candidate nuclei where single-$\beta$ decay is energetically forbidden (or sufficiently suppressed) that $\beta\beta$ decay could take place.

1.3 $\beta\beta$ decay

$\beta\beta$ decay was first investigated by Maria Goeppert-Mayer in 1935. She estimated $\beta\beta$-decay half-lifes $> 10^{17}$ yr [16]. Her work assumed that four particles were emitted, resulting in the standard $2\nu\beta\beta$ decay. This decay is shown in Fig. 1.4.

In 1939 Furry [17] was able to build off Majorana’s theory and later work by
Racah [18] to propose an alternative mode of decay: $0\nu\beta\beta$. This decay mode is possible for Majorana neutrinos via the “Racah sequence”

$$n_1 \rightarrow p_1 + e^- + \nu$$

$$\nu + n_2 \rightarrow p_2 + e^-.$$  \hspace{1cm} (1.19)

Under the Dirac theory of neutrinos, the neutrino emitted in the first step of the sequence must be a $\bar{\nu}_e$ while the neutrino absorbed in the second step is $\nu_e$. For Majorana neutrinos, the particle-antiparticle distinction is irrelevant and the process is allowed to proceed. This gives the nuclear neutrinoless double-$\beta$ decay

$$^{A}_Z X_N \rightarrow ^{A}_Z Y_{N-2} + 2e^-.$$  \hspace{1cm} (1.20)

It is immediately noted that this process violates lepton number conservation, as two leptons are produced with no antileptons. Violation of lepton number would necessitate changes to the current Standard Model and is one of the reasons why $0\nu\beta\beta$ attracts so much interest: it hints at new physics.

Furry also realized that $0\nu\beta\beta$ decay has the advantage that there are only two outgoing particles, as compared to four in $2\nu\beta\beta$ decay. This results in $0\nu\beta\beta$ having more phase space available for the decay and thus a greater transition probability. Furry noted that while $2\nu\beta\beta$ decay “could never be capable of observation because of its extremely minute probability,” this would not necessarily be the case for $0\nu\beta\beta$. Subsequent searches for $0\nu\beta\beta$ produced negative results at the level predicted by Furry and the Dirac theory of the neutrino was adopted.

Furry’s theory became obsolete when it was revealed that the weak force is left-handed. With this knowledge, it was realized that the neutrino in step one must be right-handed, while the neutrino in step two must be left-handed. As has already been stated, for massive neutrinos, handedness is not a good quantum number as it will change under a Lorentz boost. Figure 1.5 shows $0\nu\beta\beta$ decay progressing via the
“Racah sequence,” or exchange of a virtual Majorana neutrino, with a helicity flip indicated by the arrows.

Since every right-handed massive neutrino has a non-zero left-handed component the decay can proceed. The amplitude of the wrong handedness can be shown to be proportional to $m_\nu/E_\nu$, such that the neutrinoless mode is suppressed by the small neutrino mass. For a neutrino mass $O(m_{e^-})$, the neutrinoless mode would be $10^5$ times faster than the two neutrino mode. The incredibly small neutrino mass is thus responsible for $2\nu\beta\beta$ decay dominating over the neutrinoless mode, despite the neutrinoless mode having a greater phase space accessible. Under this theory, observation of $0\nu\beta\beta$ is evidence for massive Majorana neutrinos. Other mechanisms of $0\nu\beta\beta$ have been proposed, including exchange of neutralinos, gluinos, and leptoquarks, but they require new interactions or particles in addition to Majorana neutrinos [19].

Experimental evidence for $\beta\beta$ decay was first presented using geochemical techniques. This technique uses geological time scales and looks for daughter nuclei in samples of the parent isotope. The first active observation of $2\nu\beta\beta$ occurred in 1987 [20]. The experiment used a time-projection chamber to view electrons emitted from thin foils of $^{82}\text{Se}$. Of the 35 candidate isotopes for $\beta\beta$ decay, 11 of them have been observed to undergo $\beta\beta$ at the time of writing. The best candidates are selected based on their natural abundance and the decay $Q$ value. In order to produce a
signal above naturally occurring background radiation, only nuclei with a $Q$ value above 2 MeV are typically considered. The 11 nuclei that fit this criteria are summarized in Table 1.1. The phase-space factors for the $2\nu$ and $0\nu$ decay modes are also given. As was previously discussed, the $0\nu$ decay mode has a larger phase space available for decay, but the decay is suppressed by the small neutrino mass.

Experimental efforts to detect $0\nu\beta\beta$ decay have so far only produced upper limits, with one questionable claim of discovery in $^{76}$Ge [22, 23, 24]. $0\nu\beta\beta$-decay experiments measure the energy of emitted electrons. In the $2\nu$ case, the electrons and the neutrinos share the available energy, leading to a continuous electron energy spectrum up to the decay $Q$ value. In the neutrinoless mode, the electrons will take all available energy, leading to a single monoenergetic peak at the $Q$ value. The current generation of experiments search for this peak, but are also capable of measuring the $2\nu$ decay mode by observing the broad electron energy spectrum. Table 1.2 gives the best limits provided by these experiments for a variety of isotopes.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$Q_{\beta\beta}$ keV</th>
<th>nat. abund. %</th>
<th>$G_{2\nu}$ $10^{-21}$yr$^{-1}$</th>
<th>$G_{0\nu}$ $10^{-15}$yr$^{-1}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{48}$Ca</td>
<td>4272.3</td>
<td>0.187</td>
<td>15550</td>
<td>24.81</td>
</tr>
<tr>
<td>$^{76}$Ge</td>
<td>2039.1</td>
<td>7.8</td>
<td>48.17</td>
<td>2.363</td>
</tr>
<tr>
<td>$^{82}$Se</td>
<td>2995.1</td>
<td>8.7</td>
<td>1596</td>
<td>10.16</td>
</tr>
<tr>
<td>$^{96}$Zr</td>
<td>3350.4</td>
<td>2.8</td>
<td>6816</td>
<td>20.58</td>
</tr>
<tr>
<td>$^{100}$Mo</td>
<td>3034.4</td>
<td>9.8</td>
<td>3308</td>
<td>15.92</td>
</tr>
<tr>
<td>$^{110}$Pd</td>
<td>2017.8</td>
<td>11.7</td>
<td>137.7</td>
<td>4.815</td>
</tr>
<tr>
<td>$^{116}$Cd</td>
<td>2813.5</td>
<td>7.5</td>
<td>2764</td>
<td>16.70</td>
</tr>
<tr>
<td>$^{124}$Sn</td>
<td>2286.9</td>
<td>5.8</td>
<td>553.0</td>
<td>9.040</td>
</tr>
<tr>
<td>$^{130}$Te</td>
<td>2527.0</td>
<td>34.1</td>
<td>1529</td>
<td>14.22</td>
</tr>
<tr>
<td>$^{136}$Xe</td>
<td>2457.8</td>
<td>8.9</td>
<td>1433</td>
<td>14.58</td>
</tr>
<tr>
<td>$^{150}$Nd</td>
<td>3371.4</td>
<td>5.6</td>
<td>36430</td>
<td>63.03</td>
</tr>
</tbody>
</table>
Table 1.2: $\beta\beta$ decay experiments and their presented half-lifes/limits. In all cases, the first uncertainty is statistical and the second is systematic. The most recent experiments are shown for each isotope.

| Isotope | experiment | $2\nu\beta\beta$ yr $| 0\nu\beta\beta$ yr (90% C.L) |
|---------|------------|-----------------------------|
| $^{48}$Ca  | NEMO-3     | $4.4^{+0.9}_{-0.4} \pm 0.4 \times 10^{19}$ [25] | $> 1.3 \times 10^{22}$ [25] |
| $^{76}$Ge  | GERDA      | $1.84^{+0.14}_{-0.10} \times 10^{21}$ [26] | $> 2.1 \times 10^{25}$ [27] |
| $^{82}$Se  | NEMO-3     | $0.96 \pm 0.03 \pm 0.1 \times 10^{20}$ [28] | $> 1.0 \times 10^{23}$ [28] |
| $^{96}$Zr  | NEMO-3     | $2.35 \pm 0.14 \pm 0.16 \times 10^{19}$ [29] | $> 9.2 \times 10^{21}$ [29] |
| $^{100}$Mo | NEMO-3     | $7.11 \pm 0.02 \pm 0.54 \times 10^{18}$ [28] | $> 4.6 \times 10^{23}$ [28] |
| $^{130}$Te | NEMO-3     | $7.0 \pm 0.9 \pm 1.1 \times 10^{20}$ [30] | - |
| $^{130}$Te | CUORICINO  | -                           | $> 3.0 \times 10^{24}$ [31] |
| $^{136}$Xe | EXO        | $2.165 \pm 0.016 \pm 0.059 \times 10^{21}$ [32] | $> 1.1 \times 10^{25}$ [33] |
| $^{136}$Xe | KamLAND-Zen | $2.38 \pm 0.02 \pm 0.14 \times 10^{21}$ [34] | $> 1.9 \times 10^{25}$ [35] |
| $^{150}$Nd | NEMO-3     | $9.11^{+0.25}_{-0.22} \pm 0.63 \times 10^{18}$ [36] | $> 1.8 \times 10^{22}$ [36] |

1.3.1 Excited-state decays

This thesis studies decays where the daughter nucleus is left in an excited state. The excited state will then decay via $\gamma$-ray emission to the ground state. Figure 1.6 shows the double-$\beta$ decay of $^{96}$Zr to both the ground and first excited $0^+_1$ state. For double-$\beta$ decay, excited-state transitions have a smaller $Q$ value and will therefore exhibit a longer lifetime. It is noted in Ref. [37] that more research into $\beta\beta$ decays to excited states is needed from both the experimental and theoretical fronts. The theoretical motivation for these decays is discussed in Sec. 2.2.3, while the experimental motivation is discussed in Sec. 1.5

The first excited $0^+_1$ state is of particular interest because it is theoretically expected to have the largest branching ratio, excluding the ground state. The reasoning for this is discussed further in Chapter 2. It is also important to note that the $0^+_1$ state may not directly decay to the $0^+$ ground state. This is because the photon must carry helicity $\pm 1$, which requires the angular momentum of the system in the direction of radiation to change by $\pm 1$. Therefore, the $0^+_1 \rightarrow 0^+$ transition is forbidden and the decay must proceed through the intermediate $2^+$ state, producing
two $\gamma$ rays. These two coincident $\gamma$ rays form a unique experimental signature. The event topology then includes two electrons and two $\gamma$ rays, which may be used to substantially reduce the experimental background.

It should be noted that of all the nuclei observed to undergo $2\nu\beta\beta$ decay, only two of them, $^{100}\text{Mo}$ and $^{150}\text{Nd}$, have been observed to undergo $2\nu\beta\beta$ decay to the first excited $0^+_1$ state. This project began as an attempt to extend these measurements to include a third nucleus. The $Q$ value and phase-space factors for the transition to the $0^+_1$ state are given in Table 1.3. From this table, $^{96}\text{Zr}$ was selected as the best candidate due to its high phase space and $Q$ value. This agrees with systematic estimates from Ref. [37], where $^{96}\text{Zr}$ is estimated to have the second shortest half-life of all $2\nu\beta\beta$ decays to the first excited $0^+_1$ state, with $^{150}\text{Nd}$ having the shortest half-life and $^{100}\text{Mo}$ having the third shortest half-life.

Others have proposed the idea of investigating excited state decays as a consistency test for $0\nu\beta\beta$ in larger ton-scale experiments using one isotope [38]. The importance of a potential discovery often leads to the conclusion that $0\nu\beta\beta$ must be detected in multiple isotopes for proof of discovery. It is argued in Ref. [38] that it is also possible to confirm a signal using one large detector of a single isotope and investigating both the ground-state and excited-state transitions. By measuring
Table 1.3: Phase-space factors for $\beta\beta$ decay to the first excited $0^+_1$ states [21].

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$Q_{\beta\beta}(0^+_1)$ keV</th>
<th>$G_{2\nu}$ $10^{-21}$ yr$^{-1}$</th>
<th>$G_{2\nu}(0^+<em>1)/G</em>{2\nu}(0^+_{g.s.})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{48}$Ca</td>
<td>1275.0</td>
<td>0.3627</td>
<td>$2.33 \times 10^{-5}$</td>
</tr>
<tr>
<td>$^{76}$Ge</td>
<td>916.8</td>
<td>0.0698</td>
<td>$1.45 \times 10^{-3}$</td>
</tr>
<tr>
<td>$^{96}$Zr</td>
<td>2202.2</td>
<td>175.4</td>
<td>$2.57 \times 10^{-2}$</td>
</tr>
<tr>
<td>$^{100}$Mo</td>
<td>1904.1</td>
<td>60.55</td>
<td>$1.83 \times 10^{-2}$</td>
</tr>
<tr>
<td>$^{110}$Pd</td>
<td>547.7</td>
<td>0.00484</td>
<td>$3.51 \times 10^{-5}$</td>
</tr>
<tr>
<td>$^{116}$Cd</td>
<td>1056.6</td>
<td>0.8727</td>
<td>$3.16 \times 10^{-4}$</td>
</tr>
<tr>
<td>$^{124}$Sn</td>
<td>629.7</td>
<td>0.01988</td>
<td>$3.59 \times 10^{-5}$</td>
</tr>
<tr>
<td>$^{130}$Te</td>
<td>733.5</td>
<td>0.7566</td>
<td>$4.95 \times 10^{-5}$</td>
</tr>
<tr>
<td>$^{136}$Xe</td>
<td>878.8</td>
<td>0.3622</td>
<td>$2.53 \times 10^{-4}$</td>
</tr>
<tr>
<td>$^{150}$Nd</td>
<td>2631.0</td>
<td>4329</td>
<td>$1.19 \times 10^{-1}$</td>
</tr>
</tbody>
</table>

both transitions in the same experiment, systematic uncertainties can be reduced by looking at the ratio of half-lifes. This reduction in correlated errors can benefit experimental uncertainties and theoretical uncertainties in the nuclear matrix elements. Furthermore, detecting both transitions would reduce, if not completely eliminate, any unknown nuclear or radioactive backgrounds in the region of interest. An example includes $(p,n)$ reactions in the target material. The resulting nucleus will $\beta$ decay, producing a continuous electron spectrum that will overlap with the $\beta\beta$-decay signal. The exact overlap depends on the reaction $Q$ value and what, if any, nuclear states are excited in the process. This background would, however, affect the ground and excited state regions of interest differently.

1.4 Double-electron capture

Theoretical calculations for $ECEC$ were first presented in 1955 by Winter [39]. Winter noticed that the phase space available for $ECEC$ was greatly increased in the case of virtual Majorana neutrino exchange, when there are no outgoing particles. This presents a problem in that there is no mechanism to dispose of excess energy from the decay. The decay would proceed given the “monumental coincidence” [39] that an excited nuclear state exists at the correct energy to be degenerate with the
Q value of the decay. The daughter atom has some width due to the width of the excited nuclear state and width of the capturing electron states, as shown in Fig. 1.7. The total width of this state dictates the amount of degeneracy between the parent and daughter atoms and is necessary for $ECEC$ to occur. Given this degeneracy, there would be a virtual mixing of the parent atom and the daughter atom with two electron holes. This is a mixing of two atoms with different lepton number, which would only be allowed for Majorana neutrinos. As the electron holes are filled and the nucleus de-excites, the decay becomes real [40]. In the decay rate calculation, the wave function overlap between the two atoms leads to a resonant enhancement which takes the form of the Breit-Wigner formula, as will be seen in the next chapter. This process is analogous to resonant nuclear reactions and gives $0\nu ECEC$ to a degenerate state the name resonant $ECEC$. This resonant enhancement between the two degenerate states can substantially lower the half-life of the second-order decay. A 2011 search for resonant $0\nu ECEC$ candidate nuclei [41] showed that some half-lifes could be as low as $10^{22}$ yr for a neutrino mass of 1 eV. This would be a shorter half-life than $0\nu\beta\beta$ decay for an equivalent neutrino mass. For this reason, $ECEC$ is not only a possible alternative to $0\nu\beta\beta$ decay, but an experimentally feasible one, should this “monumental coincidence” occur.

The nuclear $0\nu ECEC$ process is written as

$$^{A}Z X_{N} + 2e^{-} \rightarrow ^{A}_{Z-2} Y_{N+2}^*, \quad (1.21)$$
where \( Y^* \) is an excited state of \( Y \). This process is very similar to \( 0\nu\beta\beta \) decay, in that it violates lepton number conservation and proceeds via virtual exchange of a massive Majorana neutrino, as is shown in Fig. 1.8. The main difference from \( \beta\beta \) decay is that the neutrinoless mode dominates over the \( 2\nu \) mode. For a complete degeneracy between the daughter and parent nuclei, there is no available phase space to create two neutrinos. Even if the \( Q \) value was twice the neutrino rest mass, the neutrinos would be so constricted in momentum space to disfavor the decay. In \( 0\nu\beta\beta \) experiments, one of the largest backgrounds can be the \( 2\nu\beta\beta \) spectrum, but the \( 2\nu \) process is entirely absent for \( ECEC \) between two degenerate states.

In 1983 Bernabeu et al. performed a search for candidate \( ECEC \) nuclei fitting the degeneracy criteria with a likely resonant enhancement [40]. They identified twelve candidate nuclei, with \(^{112}\text{Sn}\) being the best candidate. Bernabeu estimated \(^{112}\text{Sn}\) to have a \( 0\nu ECEC \) lifetime of \( 10^{22} \) to \( 10^{27} \) yr for a neutrino mass of 30 eV. Their work on candidate nuclei was severely limited by the available nuclear data at the time. The uncertainties on nuclear masses led to \( Q \)-value uncertainties ranging from 3 to 17 keV. More recent reviews on the topic [42] have found the experimental feasibility encouraging and noted that the process offers unique experimental advantages, should a suitable candidate nucleus be found. The \( ECEC \) field has improved in the last few years due to many high-precision mass measurements on candidate...
nuclei [43, 44]. These measurements use Penning traps to obtain sub-eV error on the $ECEC$ $Q$ value. The results of these measurements identified $^{156}$Dy as the best candidate [44], which will be the focus of the present experimental work.

The $ECEC$ process in $^{156}$Dy is shown in Fig. 1.7. The energetically forbidden intermediate nucleus $^{156}$Tb is shown, along with the excited state in the daughter $^{156}$Gd. When $^{156}$Gd deexcites, it will often produce a $\gamma$-ray cascade, with multiple $\gamma$ rays in coincidence. The experimental advantages of this signature are discussed in the next section.

1.5 Experimental technique

Excited-state decays, such as the one shown in Fig. 1.6, produce a unique signature which aids in experimental searches. Multiple $\gamma$ rays may be detected in coincidence, which substantially differentiates the decay from naturally occurring background radioactivity. For $\beta\beta$ decay, a triple coincidence between the electrons and two $\gamma$ rays can be used to effectively eliminate external backgrounds. In this decay, the energies and the angular correlation between the $\gamma$ rays are well defined.

The detection technique used in this thesis is illustrated by Fig. 1.9, which is a sketch of the two-coaxial HPGe apparatus discussed in Sec. 3.2. The sample is placed in between the two detectors, which record the two back-to-back $\gamma$ rays from the $0^+ \rightarrow 2^+ \rightarrow 0^+$ decay. The $\gamma$ rays are monoenergetic and do not experience energy degradation in the target.

An advantage of this detection technique is that the $\beta\beta$ decay electrons do not need to be detected in order to uniquely identify the decay. This allows for flexibility in the target material. Large scale experiments require either thin sources such that the electrons escape the material with their energy relatively unchanged, such as NEMO [45], or that source material to also be the detection medium, such as EXO [46] and GERDA [47]. In the present experiment, thick samples may be used and
different candidate nuclei can be investigated without changing the detector setup. The disadvantage in this approach is that the electrons are not detected, and as such the apparatus cannot distinguish $2\nu\beta\beta$ decay from $0\nu\beta\beta$.

Large scale experiments searching for $0\nu\beta\beta$ may also search for excited state decays. To take full advantage of the background reduction produced with the coincidence technique, the detectors must have excellent position reconstruction or be sufficiently segmented with little shielding in between segments. An example of the latter was provided by the CUORICINO experiment [48]. The experiment uses bolometric crystals to search of $\beta\beta$ decay in $^{130}$Te. Their search for $2\nu\beta\beta$ decay to the $0^+_1$ state investigated events with the $\beta\beta$ decay electrons depositing their energy in one crystal, while the $\gamma$ rays from the decay of the daughter nucleus are detected in adjacent crystals.
This chapter discusses the theoretical background for all of the decays studied in this work: $\beta$, $\beta\beta$, $EC$, and resonant $ECEC$. Both the two neutrino and neutrino-less modes of double-$\beta$ decay are discussed. Theoretical calculations of decay rates are shown. The result of these calculations is that a measurement of the neutrino-less decay half-life allows for a measurement of the effective neutrino mass given a theoretically calculated NME. Measurements of $2\nu\beta\beta$ to the first excited $0^+_1$ state provide an additional mode to validate and tune $0\nu\beta\beta$ NMEs, thus motivating the decay studied in this thesis. The required NMEs are discussed in detail, highlighting current theoretical progress in those calculations.

2.1 $\beta$-decay theory

The theoretical treatment of $\beta$ decay will motivate and illuminate similar features present in the $\beta\beta$-decay theory. Theory presented in this section will also apply directly to the single-$\beta$ decay of $^{96}$Zr.
2.1.1 Decay-rate calculation

Here the theoretical decay rate for $\beta^-$ decay is calculated. The calculation follows that of Ref. [49]. The decay rate may be calculated starting with Fermi’s golden rule

$$d\lambda = \frac{2\pi}{\hbar} \delta(E_f - E_i)|\mathcal{M}_{if}|^2 dN,$$  \hspace{1cm} (2.1)

where $\lambda$ is the lifetime, $\mathcal{M}_{if}$ is the matrix element for the transition between initial and final states, and $dN$ is the density of final states. The delta function enforces energy conservation and removes one degree of freedom. The density of final states consists of the electron and neutrino contributions:

$$\frac{dN_\nu}{dE_\nu} = \frac{V}{2\pi^2(h\nu)^3} \frac{(Q_\beta - E_e)^2}{(Q_\beta - E_e)^2},$$ \hspace{1cm} (2.2)

$$\frac{dN_e}{dE_e} = \frac{V}{2\pi^2(h\nu)^3} F(Z, E_e)E_e(E_e^2 - m_e^2c^4)^{1/2}.$$ \hspace{1cm} (2.3)

Energy conservation has already been enforced with $Q_\beta = E_e + E_\nu$. $F(Z, E_e)$ is the Fermi function, which accounts for the fact that the electron is not actually a free particle but will interact with the nuclear Coulomb field. The Fermi function is given by

$$F(Z, E_e) = \frac{2\pi\eta}{1 - e^{-2\pi\eta}}; \hspace{1cm} \eta = \frac{Ze^2}{\hbar v_e},$$ \hspace{1cm} (2.4)

where $v_e$ is the electron velocity. Substituting the density of states into Eq. (2.1) gives

$$d\lambda = \frac{F(Z, E)|\mathcal{M}_{if}|^2}{2\pi^3\hbar^7c^5} E_e(E_e^2 - m_e^2c^4)^{1/2}(Q_\beta - E_e)^2 dE.$$ \hspace{1cm} (2.5)

This is the differential probability per unit time for an electron emitted with energy between $E$ and $E + dE$. We can integrate this equation over all kinematically allowed energies to calculate the total decay rate

$$\lambda = \frac{f(Z, Q_\beta)|\mathcal{M}_{if}|^2}{2\pi^3\hbar^7 m^5c^4},$$ \hspace{1cm} (2.6)
where

\[ f(Z, Q_\beta) = \frac{1}{m^5 c^{10}} \int_0^{Q_\beta} F(Z, E) E(E^2 - m^2)^{1/2}(Q_\beta - E)^2 dE \quad (2.7) \]

is the dimensionless Fermi integral.

An investigation of the NME provides insight into the physics of the decay. The NME may be expanded in terms of the nuclear weak force operator \( H_\beta \) and the initial and final wave functions

\[ M_{if} = \langle \Psi_f | H_\beta | \Psi_i \rangle . \quad (2.8) \]

The final wave function is the product of the residual nucleus, the electron, and the neutrino wave functions

\[ \Psi_f = \Psi_R \Psi_e \Psi_\nu . \quad (2.9) \]

The electron and neutrino are free particles and will be represented by plane waves with normalization of \( 1/\sqrt{V} \):

\[ \Psi_e = \frac{1}{\sqrt{V}} e^{i \vec{p}_e \cdot \vec{r}/\hbar} , \quad (2.10) \]

\[ \Psi_\nu = \frac{1}{\sqrt{V}} e^{i \vec{p}_\nu \cdot \vec{r}/\hbar} . \quad (2.11) \]

These terms are multiplied together and expanded in terms of a sum over spherical waves

\[ \Psi_e \Psi_\nu = \frac{1}{\sqrt{V}} \sum_{l=0}^{\infty} \left( \frac{i}{\hbar} \right)^l (2l + 1) j_l(p_T r)| \cos \theta , \quad (2.12) \]

where \( j_l \) are the spherical Bessel functions, \( P_l \) are the Legendre polynomials, \( p_T = p_e + p_\nu \), and \( \theta \) is the angle between \( \vec{p}_e \) and \( \vec{p}_\nu \). The partial-wave expansion shows that the first term takes away \( l = 0 \) units of orbital angular momentum. These decays are called “allowed transitions.” The next term contains \( l = 1 \) units of orbital angular momentum and is known as a “first forbidden” transition. This
nomenclature continues for second forbidden transitions and so on. Each successive transition corresponds to a $\approx O(10^{-4})$ probability decrease.

Fermi did not consider spin when formulating his original theory of $\beta$ decay. This corresponds to the physical case that the electron and neutrino have opposite spins and therefore do not contribute to balancing angular momentum:

$$S_\nu + S_e = 0, \quad \text{(2.13)}$$
$$I_i = I_f + l. \quad \text{(2.14)}$$

In this event, the decays are known as Fermi transitions. The operator $H_\beta$ may be written as

$$H_\beta^F = g_V \sum_n \tau_\pm^n, \quad \text{(2.15)}$$

where $g_V$ is the Fermi constant, $\tau_\pm^n$ is the isospin raising/lowering operator, and the index $n$ sums over all nucleons. In simple shell-model calculations this sum reduces to only the valence nucleons.

The other possibility, that the neutrino and electron have parallel spins, is known as a Gamow-Teller transition. The angular momentum selection rules are as follows:

$$S_\nu + S_e = 1, \quad \text{(2.16)}$$
$$I_i = I_f + l + 1. \quad \text{(2.17)}$$

In this case, the operator $H_\beta$ must be modified to account for nuclear spin and now contains the Pauli spin operator

$$H_\beta^{GT} = g_A \sum_n \tau_\pm^n \sigma^n. \quad \text{(2.18)}$$

The coupling constant for Gamow-Teller transitions $g_A$ is fitted from experimental measurements of $\beta$ decay and found to be slightly larger than $g_V$. Typical values are around $g_A/g_V \approx 1.269$. A topic of current theoretical interest is that the effective
Table 2.1: The different transitions and angular momentum selection rules possible for $\beta$ decay

<table>
<thead>
<tr>
<th>Transition</th>
<th>$\Delta I$</th>
<th>$\Delta \pi$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Allowed</td>
<td>0, $\pm 1$</td>
<td>No</td>
</tr>
<tr>
<td>1st Forbidden</td>
<td>0, $\pm 1$, $\pm 2$</td>
<td>Yes</td>
</tr>
<tr>
<td>2nd Forbidden</td>
<td>$\pm 2$, $\pm 3$</td>
<td>No</td>
</tr>
<tr>
<td>3rd Forbidden</td>
<td>$\pm 3$, $\pm 4$</td>
<td>Yes</td>
</tr>
<tr>
<td>4th Forbidden</td>
<td>$\pm 4$, $\pm 5$</td>
<td>No</td>
</tr>
</tbody>
</table>

value of $g_A$ is quenched. The quenching arises from the limited model space in which the NME calculation is done and contributions from non-nucleonic degrees of freedom. This implies a model dependence on the value of $g_A$, which will be discussed in more detail in Sec. 2.2.3.

In general, decays are a combination of Fermi and Gamow-Teller transitions. The total matrix element is then

$$M_{if} = \left< f \left| \sum_n \tau_n^{\pm} (g_V - g_A \vec{\sigma}^n) \right| i \right>.$$  \hspace{1cm} \text{(2.19)}

This expression is reminiscent of the vector - axial vector formulation characteristic of the weak interaction, motivating writing the coupling constants as $g_V$ and $g_A$. Although only Fermi and Gamow-Teller terms are discussed here, it should be noted that additional terms arising from weak magnetism and induced pseudoscalar terms are present in the complete weak Hamiltonian. These terms are often not included in theory calculations since their effects are small.

Combining the spin of Fermi and Gamow-Teller transitions with the orbital angular momentum from Eq. (2.12), we arrive at the angular momentum selection rules and nomenclature for $\beta$ decay summarized in Table 2.1.

$^{96}$Zr differs from other $\beta\beta$ decays nuclei as single-$\beta$ decay is energetically allowed. This is shown in Fig. 2.1, where the decay is dominated by the transition to the $5^+$ state. The spin difference between $^{96}$Zr and the $5^+$ state of $^{96}$Nb requires 4 or 5 units of orbital angular momentum be taken away by the electron and neutrino, thus
Figure 2.1: (Color online) The single-β decay of $^{96}$Zr and its daughter, $^{96}$Nb. Only the three most intense decay modes in $^{96}$Mo are shown. All energies are in keV.

implying a long-lived 4th forbidden decay. QRPA techniques were used in Ref. [50] to estimate a $2.4 \times 10^{20}$ y β-decay half-life. $^{96}$Nb decays to $^{96}$Mo with a half-life of 23.35 hours. The decay will typically progress to the $5^+$ state of $^{96}$Mo as this is the allowed transition with the highest $Q$ value. As $\beta\beta$ decay will only progress to $0^+$ and $2^+$ states, this allows the decay of $^{96}$Zr by two successive β decays to be differentiated from its $\beta\beta$ decay to excited states.

2.2 $\beta\beta$ decay theory

The decay rate for double-β decay is now calculated. We first calculate the decay rate for the two neutrino case and then progress to the neutrinoless case. The NMEs are discussed in detail, as these terms contain the relevant nuclear structure quantities and are of extreme importance for extracting the effective neutrino mass from a measurement of the $0\nu\beta\beta$ half-life. In fact, improvement in the NME calculations motivates the presented measurement of $2\nu\beta\beta$ decay to the first excited state.
2.2.1 $2\nu \beta \beta$ decay-rate calculation

The $\beta\beta$ decay-rate calculation bears much similarity to the $\beta$-decay calculation. The decay rate is still calculated from Fermi’s golden rule Eq. (2.1), but as $\beta\beta$ decay is a second order weak process, the matrix element is calculated from the second order form

$$\mathcal{M}_{if} = \left| \sum_m \frac{\langle f \mid H_\beta \mid m \rangle \langle m \mid H_\beta \mid i \rangle}{E_i - E_m - p_\nu - E_e} \right|^2,$$

where $\langle m \rangle$ is the complete set of states in the intermediate nucleus. The weak Hamiltonian is a product of nuclear and leptonic currents and the final states are again a product of the final nuclear and lepton wave functions. The final state includes two neutrinos and two electrons, each with a density of states given by Eqs. (2.2) and (2.3) respectively. An additional factor of $\frac{1}{4}$ is added to account for the fact that there are two pairs of identical particles. As we are only interested in the total decay rate, the integration over all available phase space is performed. This leaves

$$\lambda^{2\nu \beta \beta} = \frac{g_\nu^4}{32\pi^7 \hbar^3} \int \frac{Q_{\beta\beta} - m_e}{m_e} F(Z, E_{e1}) p_{e1} E_{e1} dE_{e1} \int \frac{Q_{\beta\beta} - E_{e1}}{m_e} F(Z, E_{e2}) p_{e2} E_{e2} dE_{e2} \times \int_0^{Q_{\beta\beta} - E_{e1} - E_{e2}} \mathcal{M}(E_{e1}, p_{\nu_1})(Q_{\beta\beta} - E_{e1} - E_{e2} - p_{\nu_1})^2 dP_{\nu_1},$$

where $\mathcal{M}(E_{e1}, p_{\nu_1})$ is the matrix element in Eq. (2.20) and the $\delta$ function has been used to enforce conservation of energy. We have used $c = 1$ here and will continue this convention onwards. To a good approximation, the lepton energies can be replaced by their average value, that is $E_e + p_\nu \approx (M_i - M_f)/2$. This allows one to separate the nuclear matrix element from the integration over phase space, as $\mathcal{M}$ now contains
no dependence on $p_{\nu}$ and $E_e$. This result is written as

$$\lambda^{2\nu\beta\beta} = G_{2\nu\beta\beta}(Q_{\beta\beta}, Z)|M^{2\nu\beta\beta}|^2. \quad (2.22)$$

$G_{2\nu\beta\beta}$ is an exactly calculable phase-space factor given by

$$G_{2\nu\beta\beta}(Q_{\beta\beta}, Z) = \frac{g_Y^4}{32\pi^3\hbar^3} \int_{m_e}^{Q_{\beta\beta}-m_e} F(Z, E_{e1})p_{e1}E_{e1}dE_{e1} \int_{m_e}^{Q_{\beta\beta}-E_{e1}} F(Z, E_{e2})p_{e2}E_{e2}dE_{e2} \times \int_0^{Q_{\beta\beta}-E_{e1}-E_{e2}} (Q_{\beta\beta} - E_{e1} - E_{e2} - p_{\nu1})^2dP_{\nu1}. \quad (2.23)$$

The total matrix element $M^{2\nu\beta\beta}$ contains both the Fermi and Gamow-Teller contributions

$$M^{2\nu\beta\beta} = M^{2\nu\beta\beta}_{\text{GT}} - \frac{g_Y^2}{g_A^2} M^{2\nu\beta\beta}_{\text{F}}, \quad (2.24)$$

where

$$M^{2\nu\beta\beta}_{\text{F}} = \sum_m \left\langle \frac{f}{m} \sum_n \tau_+^n |m\rangle \langle m| \sum_{n'} \tau_+^{n'} |i\rangle \right\rangle, \quad (2.25)$$

$$M^{2\nu\beta\beta}_{\text{GT}} = \sum_m \left\langle \frac{f}{m} \sum_n \bar{\sigma}^n \tau_+^n |m\rangle \langle m| \sum_{n'} \bar{\sigma}^{n'} \tau_+^{n'} |i\rangle \right\rangle. \quad (2.25)$$

The NME term contains all the relevant nuclear structure effects and, as such, is difficult to theoretically calculate. This will be discussed in more detail in Sec. 2.2.3. In $2\nu\beta\beta$ decay, Fermi transitions will have all their strength transferred to the isobaric analog state in the daughter. The NME is thus dominated by the contribution from the Gamow-Teller transition. In this case, the intermediate state must be $1^+$, as all $\beta\beta$ nuclei have $0^+$ ground states.
2.2.2 $0\nu\beta\beta$ decay-rate calculation

As has already been stated, $0\nu\beta\beta$ violates conservation of lepton number and provides insight into new physics. $0\nu\beta\beta$ may occur through many non-standard model mechanisms, but they all require new particles or interactions. The most common alternative $0\nu\beta\beta$ mechanisms include the introduction of right-handed currents or heavy neutrino exchange. In this section it is assumed that this process is mediated by light Majorana neutrinos with left-handed interactions.

Once again, we begin with Fermi’s golden rule Eq. (2.1). The matrix element will contain a leptonic and nuclear part. We will first investigate the leptonic contribution. The leptonic contribution contains the weak vertex factor $\frac{1}{2}\gamma^\mu (1 - \gamma^5)$ for both lepton vertices

$$\sum_k \bar{e}(x) \gamma_\mu \frac{1 - \gamma^5}{2} U_{ek} \nu_k(x) \bar{e}(y) \gamma_\nu \frac{1 - \gamma^5}{2} U_{ek} \nu_k(y).$$

(2.26)

Here the electron neutrino state has been explicitly written as a sum of the three mass states by using the mixing matrix $U$. For Majorana neutrinos $\nu_k = \nu_k^c$ and this can be rewritten as

$$\sum_k \bar{e}(x) \gamma_\mu \frac{1 - \gamma^5}{2} U_{ek} \nu_k(x) \nu_k^c(y) \gamma_\nu \frac{1 + \gamma^5}{2} U_{ek} e^c(y).$$

(2.27)

The contraction of $\nu_k \nu_k^c$ produces the normal fermion propagator

$$\frac{i}(q^\rho \gamma_\rho + m_k)$$

$$\frac{1}{q^2 - m_k},$$

(2.28)

where $m_k$ is the neutrino mass. Substituting this into Eq. (2.27) and integrating over the momentum of the virtual neutrino gives

$$-\frac{i}{4} \int \sum_k \frac{d^4 q}{(2\pi)^4} e^{-iq(x-y)} \bar{e}(x) \gamma_\mu (1 - \gamma^5) \frac{q^\rho \gamma_\rho + m_k}{q^2 - m_k} \gamma_\nu (1 + \gamma^5) e^c(y) U_{ek}^2.$$

(2.29)
Using the uncertainty relation and the typical nucleon-nucleon spacing, the expected momentum of the virtual neutrino is \( q \sim 1/r \approx 100 \text{ MeV} \). As such, the \( m_k \) term in the denominator is negligible. Equation (2.29) can be simplified using a few easily verifiable \( \gamma_5 \) identities

\[
\{\gamma^\mu, \gamma^5\} = 0, \quad (2.30)
\]
\[
(1 - \gamma_5)(1 - \gamma_5) = 2(1 - \gamma_5), \quad (2.31)
\]
\[
(1 - \gamma_5)\gamma_\rho(1 - \gamma_5) = 0. \quad (2.32)
\]

The \( q^\mu \gamma_\rho \) term vanishes and we are left with a term proportional to the neutrino mass

\[
\langle m_{\beta\beta} \rangle \equiv \sum_k m_k U_{ek}^2. \quad (2.33)
\]

In this equation, \( m_k \) are the neutrino masses and \( U \) is the neutrino mixing matrix.

The remaining integration over the virtual neutrino momentum in Eq. (2.29) can be carried out. Using the previously discussed approximation \( m_j \ll q \) we will discard the \( m_j \) term in the denominator. First, the integration over the energy component of the \( q^\mu \) four vector is performed, leaving

\[
\frac{r}{2\pi^2} \int \frac{e^{iq\cdot\vec{r}}}{q(q + E_m - E_i + E_e)} dq. \quad (2.34)
\]

Here the substitution \( \vec{r} = \vec{x} - \vec{y} \) has been made. This term is known as the “neutrino potential.” It is contained in the NME and appears as an effect of the neutrino propagating between the two nucleons. Carrying out the integration over \( \theta \) and \( \phi \) results in the typical form of the neutrino potential

\[
H_\nu(r, E_m) \approx \frac{2R}{\pi r} \int_0^\infty \frac{\sin(qr)}{q + E_m - E_i + E_e} dq. \quad (2.35)
\]

The nuclear radius \( R \) is added to nondimensionalize the equation. The neutrino potential adds a spatial dependence of the two nucleons into the NME. This does
not appear in the $2\nu\beta\beta$ NMEs and is one of the larger differences between the two calculations. Furthermore, the inclusion of $H_\nu(r, E_m)$ into the Hamiltonian will produce spherical waves of all $l$ values. This means that both Fermi and Gamow-Teller transitions are of importance to $0\nu\beta\beta$ decay, whereas Gamow-Teller transitions dominated the $2\nu\beta\beta$ decay.

The NME is then calculated by multiplying the leptonic part, above, by the hadronic part and carrying out the transition between the initial and final states. As the neutrino potential and transition operators will depend on the intermediate energy $E_n$, we expand this expression to include the complete set of intermediate nuclear states. This will result in a NME of the form

$$\sum_m \langle f | H_{\beta} | m \rangle \langle m | H_{\beta} | i \rangle . \tag{2.36}$$

Current NMEs are typically calculated using the “closure approximation.” Given that the virtual neutrino has a relatively high energy ($\approx 100$ MeV), the intermediate state energy is simply replaced by the average value $\bar{E}$, and the sum over intermediate states $\sum_m |m\rangle \langle m|$ is replaced by unity. This removes any dependence on the intermediate state $E_m$. For example, the neutrino potential’s dependence on $E_m$ is removed, although this is a reasonable approximation since $q \gg E_m$.

Next, the integration over all available phase space is performed. The closure approximation allows the nuclear structure terms to be separated from the exactly calculable phase-space integral. The result is that the lifetime is now a product of three separate factors:

$$\lambda^{0\nu\beta\beta} = G_{0\nu\beta\beta}(Q_{\beta\beta}, Z) |M^{0\nu\beta\beta}|^2 \langle m_{\beta\beta} \rangle^2 . \tag{2.37}$$

The final NME $M^{0\nu\beta\beta}$, in the closure approximation, is given by

$$\left\langle \sum_{n,n'} H_\nu(r, \bar{E}) \tau_+ \tau_+ [h_F(q) + h_{GT}(q) \vec{\sigma}_n \cdot \vec{\sigma}_{n'} + h_T(q) S_{nn'}(q)] i \right\rangle , \tag{2.38}$$
where the tensor operator

\[ S_{nn}(q) = 3[(\vec{\sigma}_n \cdot \hat{q})(\vec{\sigma}_{n'} \cdot \hat{q})] - \vec{\sigma}_n \cdot \vec{\sigma}_{n'} \]  

(2.39)

The parameters \( h_F(q) \), \( h_{GT}(q) \), and \( h_T(q) \) contain the higher order corrections from weak magnetism and induced pseudoscalar terms in the weak nucleon current. If these corrections are ignored, the parameters reduce to the previously discussed constants \( g_V^2 \), \( g_A^2 \), and \( h_T(q) = 0 \) [19]. The total neutrinoless NME is written as

\[ M^{0\nu\beta\beta} = g_A^2 \left[ M_{GT} - \left( \frac{g_V}{g_A} \right)^2 M_F + M_T \right] \]  

(2.40)

where \( M_T \) is the contribution to the matrix element from the tensor operator \( S_{nn} \).

The phase-space factor \( G_{0\nu\beta\beta} \) is given by

\[ G_{0\nu\beta\beta}(Q_{\beta\beta}, Z) = \frac{1}{4\pi^3 \hbar^3} \int F(Z, E_{e_1})P_{e_1}E_{e_1}dE_{e_1} \int F(Z, E_{e_2})P_{e_2}E_{e_2} \]

\[ \times \delta(Q_{\beta\beta} - E_{e_1} - E_{e_2})dE_{e_2} \]  

(2.41)

Using a few approximations for the Fermi function, this integral and the one in Eq. (2.23) may be performed analytically. This results in \( G_{2\nu\beta\beta} \sim Q_{\beta\beta}^{11} \) and \( G_{0\nu\beta\beta} \sim Q_{\beta\beta}^5 \). As expected, the two-body decay mode is preferred over the four-body decay mode when only phase space is taken into account.

The result in Eq. (2.37) shows the importance of accurate NME calculations. An experimental measurement of the \( 0\nu\beta\beta \) lifetime would allow for a measurement of the neutrino mass \( m_{\beta\beta} \). This requires the phase-space factor and NME to be theoretically calculated. The phase-space integral may be done using simple computational techniques. The NMEs, on the other hand, requires extensive nuclear structure theory and state of the art computational models. This is discussed in the following section.
2.2.3 \( \beta\beta \) Nuclear Matrix Elements

Here the evaluation of NME is discussed in more detail. The closure approximation was already discussed in the calculation of the \( 0\nu\beta\beta \) NME. This technique may also be applied to the two-neutrino case, and the NME from Eq. (2.25) are now written as

\[
M_{2\nu\beta\beta}^F = \frac{1}{2}(Q_{\beta\beta} + 2m_e c^2 + E_{0_i} - E_i),
\]

\[
M_{2\nu\beta\beta}^{GT} = \frac{1}{2}(Q_{\beta\beta} + 2m_e c^2 + E_{1_i} - E_i).
\]

The validity of this approximation in \( 2\nu\beta\beta \) has been questioned in some works [51]. This makes the calculation of \( 2\nu\beta\beta \) decay NME more challenging as the nuclear structure of the intermediate state must be included. A common alternative, known as the single-state dominance hypothesis (SSD), suggests that the decay is dominated by the lowest \( 1^+ \) intermediate state for Gamow-Teller transitions, or the lowest \( 0^+ \) state for Fermi transitions. Under the SSD hypothesis, Eq. (2.25) is written as

\[
M_{2\nu\beta\beta}^F = \frac{\langle 0_f^+ | \sum_{nm} \tau_+^n \tau_+^{n'} | 0_i^+ \rangle}{\frac{1}{2}(Q_{\beta\beta} + 2m_e c^2 + E_{0_i} - E_i)},
\]

\[
M_{2\nu\beta\beta}^{GT} = \frac{\langle 0_f^+ | \sum_{nm} \tau_+^n \tau_+^{n'} \vec{\sigma}^n \cdot \vec{\sigma}^{n'} | 0_i^+ \rangle}{\frac{1}{2}(Q_{\beta\beta} + 2m_e c^2 + E_{1_i} - E_i)}.
\]

If the SSD hypothesis is correct, the closure approximation is clearly not valid for \( 2\nu\beta\beta \) NMEs since only \( 0^+ \) and \( 1^+ \) states from the intermediate nucleus contribute.

The main advantage of the SSD hypothesis and closure approximation is the ability to decouple the phase-space factor and the NME, as shown in Eq. (2.22). The other alternative, calculating the transition for each excited \( 0^+_N \) and \( 1^+_N \) state in the intermediate nucleus and summing the results, represents a large theoretical
undertaking. Furthermore, the existence of the giant Gamow-Teller resonance, with a width of 5 to 10 MeV, in the intermediate nucleus is often not included in the model space [51], but its inclusion would greatly increase the difficulty of the calculations without the closure or SSD approximations.

Evaluation of NME using microscopic models

The evaluation of NMEs is presently a very challenging task in the nuclear theory community. The full solution involves effectively solving the nuclear many body problem. Figure 2.2 shows a collection of $0\nu\beta\beta$ NME calculations using different mean-field techniques. These include the quasiparticle random phase approximation (QRPA), interacting shell model (ISM), microscopic interacting boson model (IBM-2), projected Hartree-Fock-Bogoliubov (PHFB), and density functional theory (DFT). Among them, QRPA and ISM are the two most widely used nuclear models for the $A \sim 40 - 140$ mass range. It is customary to multiply NMEs by the nuclear radius $R = 1.2A^{1/3}\text{ fm}$ in order to achieve dimensionless NMEs for direct comparisons. The large variation between models is immediately apparent. It is not known how to assess uncertainties or assign accuracy to the different calculations. This uncertainty will be directly transferred into uncertainties on the effective neutrino mass, should a measurement be made. Models are often assessed by their ability to reproduce the $2\nu\beta\beta$-decay rate. As such, accurate $2\nu\beta\beta$-decay data are necessary to evaluate theoretical NMEs.

The NME calculations should only be compared to other calculations for the same nuclei. A complete understanding of these theories is too long for this work, but the basic principles, merits, and issues concerning each model will be discussed.

QRPA uses phenomenological interactions with free parameters fitted to experimental results. The most notable of these parameters is the interaction strength of the particle-particle force $g_{pp}$. NME calculations performed using QRPA have a
high sensitivity on $g_{pp}$ and realistic values of $g_{pp}$ often lead to solutions far from the unperturbed ground state, where QRPA is not fully applicable [56]. In $0\nu\beta\beta$ decay, the value of $g_{pp}$ is usually tuned to reproduce the $2\nu\beta\beta$-decay lifetimes. Because of this, QRPA can not simultaneously reproduce $2\nu\beta\beta$ and single-$\beta$ decay rates [19]. It would be possible to tune $g_{pp}$ using both the ground-state and excited-state decay in order to achieve more sensible values, should the excited-state decay be measured.

The range of QRPA NMEs (as shown by the error bars in Fig. 2.2) include the uncertainty on $g_{pp}$ as determined from the uncertainty of experimental measurements of the $2\nu\beta\beta$ lifetime. This range also includes effects from using different sets of single-particle states, different values of $g_A$, and two different treatments of short-range correlations. The calculations were all done in [52] and illustrate variations within QRPA methodology.

The shell model is one of the oldest nuclear models, dating back to 1949 [49]. The original shell model assumed that nucleons were independent and bound to a central potential, much like atomic electrons. Additional terms were added to account for...
spin-orbit interactions. Nucleon-nucleon and pairing interactions are added in the ISM and are necessary to calculate $\beta\beta$-decay NMEs. The ISM is expected to produce the most reliable results for light, closed-shell nuclei, such as $^{48}$Ca. Deformed nuclei, such as $^{150}$Nd, require shell-model calculations to contain ad hoc corrections and are typically not regarded as accurate.

Comparing the shell model and QRPA methodologies illuminates the differences in approach. Shell-model calculations include far fewer single-particle states, which is compensated for by including complex correlations not included within the QRPA framework. This allows shell-model calculations to reproduce complete spectroscopic data for most nuclei. QRPA, on the other hand, is limited to only reproducing ground state transitions as correlations necessary for two-particle excitations are not included [19]. Lastly, it should be noted that ISM calculations are generally considered more challenging than QRPA calculations.

The IBM has only recently been applied to $\beta\beta$ matrix element calculations. The complete methodology is discussed in Ref. [57]. The model maps pairs of shell-model states to bosonic wave functions. This removes the antisymmetrization procedure in order to ease interacting particle calculations. The fermion operator $H$ is also mapped onto a boson space in order to complete the NME calculation. The mappings are only done to leading order and next to leading order contributions are shown to be negligible [57].

A recent publication [58] using the IBM showed promise for improved $0\nu\beta\beta$ NMEs to excited states. Transition strengths between nuclear states of $^{154}$Gd were measured and shown to be very sensitive to the proton-neutron interaction strength used in the IBM framework. Using this updated interaction strength, new calculations of the $0\nu\beta\beta$ NME for $^{154}$Sm to the first excited $0^+_1$ state in $^{154}$Gd showed a larger NME than previously thought. The authors hypothesized that a larger effect would be observed in $^{150}$Nd. The work depended on the nuclear deformation of $^{154}$Sm and
$^{150}$Nd, and as such cannot be applied to all $\beta\beta$-decay candidates.

The PHFB method calculates NMEs with nuclear wave functions found using a modified Hartree-Fock method. This procedure separates the nuclear Hamiltonian into a mean-field and two-body potential. The starting wave functions are found using only the mean-field potential. The pairing energy is then calculated using these wave functions. This pairing energy is added into the original Hamiltonian, which can once again be solved for nuclear wave functions. Multiple iterations of this technique will converge to a nuclear wave function, which is then used to calculate the NME. Strengths of this method include that it accurately reproduces pairing and deformation degrees of freedom in the $94 \leq A \leq 150$ mass region. It is not possible, however, to study odd-odd nuclei using PHFB as applied in [54]. For this reason $\beta$ decay and the intermediate nucleus during $\beta\beta$ decay cannot be studied using PHFB. This means that the closure approximation must be used for these NME calculations, and that $2\nu\beta\beta$ NMEs may not be calculated, as the SSD hypothesis requires a theoretical description of the odd-odd intermediate nucleus.

The DFT method began as an alternative to Hartree-Fock with decreased computation times. The method assumes a density-dependent effective two-body interaction between the nucleons. Another improvement from PHFB is the addition of shape mixing when calculating deformation effects. This is used to solve the quantum many-body problem and self-consistently calculate nuclear properties, such as nuclear deformation, transitions, and decay rates. Comparatively, a method such as QRPA has several free parameters which must be adjusted independently for each nuclear property calculation, even within the same nucleus. The advantages of DFT are clearly the internal consistency of calculations (especially when corrections from pairing and deformation are of interest) and fast computation times. The faster computation times allow the DFT calculations to include a larger configuration space than the PHFB calculations. DFT, however, suffers from the same drawback as
PHFB: it is not possible to compute odd-odd nuclei within the theory.

Quenching of $g_A$ was previously mentioned in regards to $\beta$ decay. This phenomena has also been proven to play a role in $\beta\beta$ decay. It is known that the quenching is model dependent. Extracting $g_A$ from experiments gives $g_{A,eff} = 1.269A^{-\gamma}$, where $\gamma_{IBM-2} = 0.18$ and $\gamma_{ISM} = 0.12$. It is thus important to compare theoretical $2\nu\beta\beta$ NMEs to experimental measurements to determine the effects of quenching. By measuring the $2\nu\beta\beta$ to an excited state, additional information is given on the role of the quenching of $g_A$ and more accurate values may be obtained.

*Nuclear Matrix Elements to excited final states*

It has already been stated that accurate data on the $2\nu\beta\beta$ mode is needed to validate NME calculations. By measuring the decay to the first excited $0^+$ state, additional criteria is provided for validation. Furthermore, the excited state decay mode may be used to tune NME calculations, as has already been seen with $g_{pp}$ in QRPA.

Transitions to excited final states insert additional complications because the excited state must also be accurately modeled in the theory, although this is often easily done. For $0\nu\beta\beta$ decay, the electrons are preferentially emitted in a $s_{1/2}$ wave, and spin-0 states are the most likely final states. Excited $2^+$ states require higher order terms in the electron partial-wave expansion. In this work we investigate the $0^+_1$ state, which is a two-quadrupole phonon state produced by nuclear vibrations. The $2^+$ state appearing in the $0^+_1 \rightarrow 2^+ \rightarrow 0^+_{g.s.}$ decay results from the rotational band.

Figure 2.3 shows $2\nu\beta\beta$ NME calculations to both the $0^+_{g.s.}$ and $0^+_1$ states using the IBM. This calculation was done using the closure approximation for both the ground and excited states. This makes the results directly comparable to $0\nu\beta\beta$ decay NMEs, however, the NME may not be accurate as the closure approximation is assumed to be invalid for $2\nu\beta\beta$ decay. As has already been mentioned, the SSD hypothesis would
likely produce more reliable results. This calculation shows that $^{96}\text{Zr}$’s decay to the $0^+_1$ state is largely suppressed compared to other nuclei. A possible explanation for the suppression is that $^{96}\text{Zr}$ is closed shell in protons and neutrons. $^{96}\text{Zr}$ contains 40 protons, filling the $2p_{1/2}$ shell, and 56 neutrons, filling the $2d_{5/2}$ shell. It would be very interesting to see how this calculation compares to one done using the ISM, which would better model the shell closure of $^{96}\text{Zr}$. Unfortunately, since most nuclear models normalize decay rate parameters to the $2\nu\bar{\beta}\beta$ decay, the IBM is the only one to publish NME for $2\nu\bar{\beta}\beta$ to the ground and excited states.

More NME calculations for $\beta\beta$ decay to excited states have been performed for the neutrinoless case. The ratio of the ground state to excited state NME are shown in Fig. 2.4. A large variation is shown in the ratio of NMEs. Interestingly, doubly magic $^{48}\text{Ca}$ is the only nucleus to show an increase in the NME to the excited state transition. For the QRPA calculations in Ref. [60], two different techniques were used to model the excited state transition: the recoupling method (RCM) and the boson expansion method (BEM). The RCM was an older method that recoupled the two $pn$ operators into a $pp$ and $nn$ operator. This method is disadvantageous in that
it includes nonphysical contributions. This issue is fixed in the BEM, where nuclear operators are expanded as polynomials under a bosonic mapping and then evaluated. The BEM is not without its disadvantages, though, including a strong dependence on the strength of the two-body interaction.

2.3 \textit{ECEC} theory

The theory of double-electron capture is now discussed. Specifically, the resonantly enhanced $0\nu\beta\beta$ decay mode will be investigated. The decay-rate formula is derived, including the enhancement factor resulting from the degeneracy. Lastly, recent NME calculations of the \textit{ECEC} transitions are presented.

2.3.1 \textit{EC} decay-rate calculation

As a short aside, \textit{EC} decay is discussed and the decay rate calculated. The decay rate is calculated from Fermi’s golden rule and is very similar to the $\beta$-decay rate calculation. The produced neutrino has the same density of states as Eq. (2.2). As this is now a one-body decay, conservation of energy forces the neutrino to be
monoenergetic and the integrals over phase space become trivial. The initial state wave function is now the product of the initial nucleus and the capturing electron’s wave function. This depends on the orbital of the capturing electron $X$. For electron capture this is simplified to the atomic electron wave function evaluated at the origin $|\Psi_X(0)|$. Substituting into Eq. (2.1), results in

$$
\lambda^e = \frac{1}{\pi \hbar^2 c^3} E_\nu^2 |M_{ij}|^2 |\Psi_X(0)|^2.
$$

(2.44)

An additional factor of two is added to account for the fact that two electrons occupy shell $X$. This rate contains many of the same features as the $\beta$-decay rate, including a NME. The remaining terms will be known as prefactors in order to distinguish them from the phase-space factors of $\beta$ decay.

### 2.3.2 ECEC decay-rate calculation

As expected, the calculation of the $0\nu ECEC$ decay rate is very similar to that of $0\nu \beta\beta$ decay. The process will be mediated by the exchange of a massive virtual neutrino as shown in Fig. 1.8. This results in a nearly identical leptonic contribution to the nuclear matrix element. The main difference is that the free electron wave functions are now replaced by the orbital electron wave functions $\Psi_X$ and $\Psi_Y$. Substituting these wave functions into Eq. (2.29) and performing the same simplifications results in

$$
\mathcal{M}^{0\nu ee} = 2 \sum_k \left( \frac{g_A}{\sqrt{2}} \right)^2 \langle i | H_\beta^0 m_k U_{e_k}^2 H_\nu(r, E) H_\beta^\sigma | f \rangle \times \Psi_X \gamma_\rho (1 - \gamma_5) \gamma_\sigma \Psi_Y.
$$

(2.45)

The closure approximation has been used to remove the sum over intermediate states. As expected, this equation contains all the same terms: two weak operators $H_\beta$, the neutrino potential $H_\nu$, and the effective neutrino mass $m_k U_{e_k}^2$. Note that the weak operator $H_\beta$ now contains the isospin lowering operator $\tau_-$. The electron
wave functions will later be separated into the prefactor term and the remainder of Eq. (2.45) will become the NME.

Given a degeneracy between the initial and final atoms, a resonant enhancement can occur in the rate. This has been studied using different methods, but here we will use the argument of Ref. [41]. The two mass degenerate atoms of differing lepton number are mixed by the weak interaction, resulting in oscillations between the two atoms. These oscillations only become real once the valence electron holes are filled and the nucleus de-excites. This system is analogous to two coupled oscillators, one of which experiences friction. This can be modeled phenomenologically using a $2 \times 2$ Hamiltonian

$$H_{\text{eff}} = \begin{pmatrix} M_i & V \\ V^* & M_f - \frac{i}{2} \Gamma_{XY} \end{pmatrix}.$$ \hfill (2.46)

$M_i$ and $M_f$ are the initial and final nuclear masses. Note that the final mass includes the binding energy of the two capturing electrons and the energy of the excited state: $M_f = M_{f0} + B_{XY} + E^*$. The final state has a width $\Gamma_{XY}$ which is the sum of the width of the two capturing electrons and the excited nuclear state

$$\Gamma_{XY} = \Gamma_X + \Gamma_Y + \Gamma^*.$$ \hfill (2.47)

The off-diagonal term $V$ produces weak mixing of the atoms, which is permitted for Majorana neutrinos and violates lepton number conservation.

Diagonalizing $H_{\text{eff}}$ results in two eigenvalues

$$\lambda_+ = M_i + \Delta M - \frac{i}{2} \Gamma_1,$$ \hfill (2.48)

$$\lambda_- = M_f - \frac{i}{2} \Gamma - \Delta M + \frac{i}{2} \Gamma_1.$$ \hfill (2.49)
where
\[ \Delta M = \frac{V^2(M_i - M_f)}{(M_i - M_f)^2 + \frac{1}{4}\Gamma_{XY}^2}, \quad (2.50) \]
\[ \Gamma_1 = \frac{V^2\Gamma_{XY}}{(M_i - M_f)^2 + \frac{1}{4}\Gamma_{XY}^2}. \quad (2.51) \]

Using typical numerical values, the imaginary parts of \( \lambda_\pm \) are negative and the states therefore decay with amplitude proportional to \( \Gamma_1 \). This produces the expected Breit-Wigner formula for the resonant enhancement of the decay
\[ EF = \frac{\Gamma_{XY}}{\Delta^2 + \frac{1}{4}\Gamma_{XY}^2}, \quad (2.52) \]
where \( \Delta = Q - B_{XY} - E^* \). The Breit-Wigner enhancement factor will appear in the final decay-rate calculation.

Combining this result with the NME found in Eq. (2.45) and substituting into Fermi’s golden rule gives the final lifetime calculation. This is traditionally broken into four factors: the prefactor \( G^{0\nu}_{\nu ee} \), the NME \( M^{0\nu ee} \), the neutrino mass, and the enhancement factor. The prefactor contains the dependence on the orbital electron-nucleus overlap \( |\Psi_X|^2|\Psi_Y|^2 \) and all additional constants. The total decay rate is then
\[ \lambda^{0\nu ee} = g_A^4 G^{0\nu}_{\nu ee} |M^{0\nu ee}|^2 (m_\nu)^2 \frac{\Gamma_{XY}}{\Delta^2 + \frac{1}{4}\Gamma_{XY}^2}. \quad (2.53) \]

Once again, there is a Fermi, Gamow-Teller, and tensor contribution to the NME, identical to Eq. (2.40). The computation of NMEs is identical to Eq. (2.38) but with the isospin raising operator \( \tau_+ \) replaced by \( \tau_- \).

It was previously noted that \( 0\nu ECEC \) will dominate over the two neutrino mode. An estimate of the ratio of \( \Gamma^{0\nu}/\Gamma^{2\nu} \) is done in [42]. This ratio has a large dependence on the degree of degeneracy \( \Delta \). Using a neutrino mass of 1 eV, the ratio peaks at 10^9. Even with a \( \Delta \) of 1 keV, \( \Gamma^{0\nu}/\Gamma^{2\nu} \) is still a very favorable 10^4. The ratio continues to
Table 2.2: The possible excited states in the daughter, $^{156}$Gd, after $0\nu ECEC$ of $^{156}$Dy. The physical state in the daughter has energy $E^\ast$. Electrons are captured from the $XY$ orbitals with a total binding energy $B_{XY}$. The degree of degeneracy for the decay is given by $\Delta = Q - E^\ast - B_{XY}$, where $Q$ is the $ECEC$ $Q$ value. For each transition there is an enhancement factor in the rate, $EF$, and a wave-function overlap for the atomic electrons and the nucleus in atomic units, $|\Psi_X|^2|\Psi_Y|^2$.[44].

| $E^\ast$ (keV) | $I^\pi$ | $e^-$ orbitals (XY) | $B_{XY}$ (keV) | $\Delta$ (keV) | $\Gamma_{XY}$ (eV) | $EF$ | $|\Psi_X|^2|\Psi_Y|^2$ |
|----------------|--------|---------------------|----------------|--------------|---------------|------|------------------|
| 1946.375       | 1$^-$  | KL$_1$              | 58.822 (8)     | 0.75 (10)    | 26            | $4.1 \times 10^6$ | 1.23 $\times 10^{10}$ |
| 1952.385       | 0$^-$  | KM$_1$              | 52.192 (8)     | 1.37 (10)    | 35            | $1.7 \times 10^6$ | 2.68 $\times 10^9$   |
| 1988.5         | 0$^+$  | L$_1$L$_1$          | 16.914 (8)     | 0.54 (24)    | 8             | $2.5 \times 10^6$ | 1.65 $\times 10^9$   |
| 2003.749       | 2$^+$  | M$_1$N$_3$          | 2.160 (24)     | 0.04 (24)    | 15            | $7.7 \times 10^8$ | 1.52 $\times 10^4$   |

The transition probability is the product of these two factors, and the four states are listed in order of most to least likely. Within the experimental error of these measurements, the 2003.7 keV state has the possibility of being completely degenerate. In this case, the enhancement factor is $2.4 \times 10^{10}$. This state, however, requires the capturing of M$_1$ and N$_3$ shell electrons, which is very unlikely, as shown in column eight. The 1988.5 keV state suffers from a dearth of nuclear data. The branching ratios from this 0$^+$ state and the properties of this excitation are unknown.

2.3.3 $0\nu ECEC$ NME calculations

A number of theory groups have recently started publishing $ECEC$ NME calculations. Once again, models such as energy density functional method (EDFT) [61], QRPA [41], and the IBM [62] are used. NME calculations are more difficult for resonant $0\nu ECEC$ than for $0\nu\beta\beta$ because the excited state energies and wave functions decrease as $\Delta$ increases, but candidate nuclei with larger $\Delta$ are expected to exhibit long lifetimes due to the smaller enhancement factor.
Table 2.3: The $0\nu ECEC$ NME for $^{156}$Dy. The NME is given for the transition to the first five $0^+$ states. The NMEs are broken up into the three different contributions, as given by Eq. (2.40). The 1988 keV state of interest for resonant $0\nu ECEC$ is $0^+_4$.

<table>
<thead>
<tr>
<th>State</th>
<th>$M^{0\nu}_{GT}$</th>
<th>$M^{0\nu}_F$</th>
<th>$M^{0\nu}_T$</th>
<th>$M^{0\nu}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$0^+_1$</td>
<td>2.796</td>
<td>-0.398</td>
<td>0.132</td>
<td>3.175</td>
</tr>
<tr>
<td>$0^+_2$</td>
<td>1.532</td>
<td>-0.227</td>
<td>0.076</td>
<td>1.749</td>
</tr>
<tr>
<td>$0^+_3$</td>
<td>0.403</td>
<td>-0.065</td>
<td>0.022</td>
<td>0.466</td>
</tr>
<tr>
<td>$0^+_4$</td>
<td>0.265</td>
<td>-0.048</td>
<td>0.017</td>
<td>0.311</td>
</tr>
<tr>
<td>$0^+_5$</td>
<td>0.302</td>
<td>-0.046</td>
<td>0.016</td>
<td>0.346</td>
</tr>
</tbody>
</table>

must also be evaluated. Furthermore, resonant $0\nu ECEC$ candidates are frequently heavy and strongly deformed nuclei, which requires additional theoretical understanding.

The NME for $^{156}$Dy was only calculated by [62], who only calculated the NME for $0^+$ transitions. A summary of these results is shown in Table 2.3. The $0^+_4$ state is the 1988 keV state of interest for the resonant $0\nu ECEC$. The NMEs show a trend of decreasing as higher excited states are considered, which disfavors transitions to excited nuclear states. Indeed, [62] calculates $T_{1/2} = 2.89 \times 10^{30}$ yr for $\langle m_\nu \rangle = 1$ eV. These results were published in 2014, after the present search for the resonant $0\nu ECEC$ decay was already underway. These theoretical lifetimes for $0\nu ECEC$ are of the same order as those obtained for $\beta^+\beta^+$ decay [63, 64]. Both lifetimes are considerably longer than those expected for $0\nu\beta^-\beta^-$.

$0\nu ECEC$ NMEs were also calculated for three other nuclei: $^{152}$Gd, $^{164}$Er, and $^{180}$W. For these three nuclei, the IBM calculations of [62] are comparable (the same order of magnitude) as the QRPA calculations. The EDFT NMEs [61], however, are close to one order of magnitude smaller than these two calculations. The reason for this discrepancy is unknown [62].
This chapter discusses the experimental apparatuses used and the physics motivating their use. The detector properties, electronics, data-acquisition system, and shielding are all discussed. In addition, a review of previous experimental searches is given, highlighting the experimental differences and improvements in the present work.

3.1 $\gamma$-ray detection

The work presented in this thesis is based on the detection of $\gamma$ rays. Electrons produced by naturally occurring radioactivity do not have sufficient energy to penetrate the 1-2 mm aluminum or magnesium shell surrounding the detectors and produce a signal. This includes the electrons produced from $\beta\beta$ decay, which will deposit all their energy in the sample or sample container. Electrons produced in or on the surface of the detectors, however, will be detected with near 100\% efficiency. In fact, $\gamma$ rays are detected in this way, by being converted into electrons or positrons inside the active detector volume.

$\gamma$ rays primarily interact with matter through one of three processes: the photoelectric effect, Compton scattering, and pair production. The photoelectric effect is
the dominant process below \(\approx 140 \text{ keV} \ [65]\). In this process, the full energy of the \(\gamma\) ray is transferred into an atomic electron, less the electron’s binding energy. For the detectors and energies considered here, the electron can be assumed to deposit its full energy into the detector via Coulomb scattering. Pair production occurs when the \(\gamma\) ray converts into an electron-positron pair. This process is only energetically allowed for \(\gamma\) rays above 1.022 MeV (the sum rest mass of the electron and positron) and will become the dominate process above \(\approx 8 \text{ MeV} \ [65]\). Once again, the electron can be assumed to deposit its full energy in the detector. The positron deposits its kinetic energy, slowing down in the detector via Coulomb scattering, then annihilates with an electron to form two back-to-back 511 keV \(\gamma\) rays. In order for the full energy to be deposited in the detector, these 511 keV \(\gamma\) rays must also be detected. It is common for one or both of these \(\gamma\) rays to escape the detector volume, leaving a single escape peak 511 keV below the \(\gamma\) ray’s energy or a double escape peak 1022 keV below the \(\gamma\) ray’s energy. Compton scattering dominates the median energies between the photoelectric effect and pair production. The \(\gamma\) ray scatters off an atomic electron, imparting part of its energy to the electron and deflecting at some angle. The electron will again deposit its full energy in the detector. In order to detect the full energy of the incident \(\gamma\) ray, we require Compton scattering (or multiple Compton scatters) followed by photoelectric absorption. A large continuous background is created by Compton scattered \(\gamma\) rays escaping the detector, only having deposited part of their energy.

High-energy electrons in the detector will produce multiple charge carriers (such as electron-hole pairs) as they lose their energy via Coulomb scattering. The number of pairs \(N\) is proportional to the energy of the incident electron divided by the electron-hole pair separation energy of the material. Efficient collection of all charge carriers will produce a signal proportional to the incident energy. This gives rise to an important detector characteristic: the detector response is linear with the incident
energy. An estimate of the detector’s energy resolution is made by assuming the creation of charge carriers is a Poisson process with standard deviation $\sqrt{N}$. In this case, the detector’s resolution $R$ is

$$R = \frac{FWHM(E_0)}{E_0} \approx \frac{2.34}{\sqrt{N}}. \quad (3.1)$$

An additional term, called the Fano factor, is usually added to account for the fact that the charge carriers are not produced independently and therefore are not a true Poisson process [66]. In the case of semiconductor detectors, this further increases the resolution. Regardless, materials with smaller band gaps result in higher resolution detectors. For this reason semiconductor detectors exhibit better energy resolution than scintillator detectors. In germanium, discussed below, only 2.96 eV is required to form an electron-hole pair and thousands of charge carries are produced for a typical keV-scale $\gamma$-ray interaction.

### 3.1.1 HPGe detectors

HPGe detectors have become the standard for high precision $\gamma$-ray spectroscopy. This is mainly because of their excellent energy resolution, around 0.2% FWHM for 1 MeV $\gamma$ rays. They are a semiconductor detector that measures the energy deposited in the active volume by collection of electron-hole pairs. Both experimental apparatuses use HPGe detectors as their basis. An example of a background HPGe spectrum over the energy range 200 keV to 2 MeV is shown in Fig. 3.1. Note the excellent energy resolution illustrated by distinct background $\gamma$ transitions.

Semiconductors are characterized as $n$ type if they contain donor impurities (here, lithium), which contribute additional electrons. $p$-type semiconductors contain acceptor impurities (here, boron), contributing additional holes. The active detector region is formed at a $pn$ junction. A depletion region forms at this junction when holes from the $p$-type crystal combine with electrons from the $n$-type crystal. This
Figure 3.1: (Color online) The spectra from one coaxial HPGe. The reduction in background from the NaI veto and $\gamma - \gamma$ coincidence cuts are shown.

Figure 3.2: (Color online) Schematic of a coaxial $p$-type HPGe detector. The two regions of the diode, the reverse bias, and electron-hole mobility are all labeled. Heavily doped regions are labeled with a ‘+’ superscript and high-purity regions are labeled with Greek lettering. Commonly, one region is doped more heavily such that the depletion region extends further into the purer region.
It can be shown [65] that the thickness of the depletion region $d$ is given by

$$d = \left( \frac{2\epsilon V}{eN} \right)^{1/2},$$

(3.2)

where $\epsilon$ is the dielectric constant, $V$ is the reverse bias voltage, $e$ is the electron charge, and $N$ is net impurity concentration. For the depletion region to span the entire detector volume, with applied voltages on the order of 1 kV and dimensions representative of germanium detectors, the impurity concentration must be less than 1 part in $10^{12}$. This incredibly low dopant concentration motivates the name “High-Purity Germanium” (HPGe). Furthermore, the required purity level of these detectors makes them excellent detectors for low background physics, since they contain very little naturally occurring radioactivity.

The ability to produce germanium with the required dopant concentration was not developed until the mid-1970s [65], when a technique known as zone refining was developed. Electronics-grade germanium is heated to its melting point ($938 \degree C$) using radio-frequency (RF) heaters. As the germanium solidifies, impurities are left in the liquid phase. By sweeping the RF heaters through a length of germanium, the impurities migrate towards the end of the crystal, which is then discarded. After many passes, a factor of $>100$ reduction in the impurity concentration is achieved [67]. Ultra-pure germanium is then melted and a small crystal of germanium is slowly pulled through. The liquid germanium crystallizes onto the seed, with the seed crystal’s speed determining the crystal size. The final crystal is then precision machined into the final shape, such that the crystalline structure is not disturbed. The coaxial detector shape is chosen to optimize the detector’s efficiency for complete energy deposition of $\gamma$ rays. Planar geometries are not thick enough for higher energy $\gamma$ rays to efficiently deposit all their energy.

The detector edges, the region furthest from the inner contact, often suffer from a
low electric field. Interactions in these low field regions result in pulses with long rise times and incomplete charge collection [67]. The detector edges are thus bulletized to remove issues caused by the low field region. Finally, the $p^+$ region is created by ion implanting boron atoms into the first 0.3 microns of the crystal. Lithium is diffused into the first 700 microns of the surface to create the $n^+$ region.

HPGe detectors must be cooled in order to minimize electronic noise from thermal fluctuations. Typically, liquid nitrogen (77 K) is used as a coolant. The germanium crystals are installed inside a vacuum shell and connected to a liquid nitrogen dewar. A copper “cold finger” cools the detector by being in thermal contact with both the crystal and liquid nitrogen.

Manufacturers couple HPGe detectors with a charge-sensitive preamplifier. This circuit collects charge from the detector and forms a voltage pulse suitable for further electronic processing. The preamplifier usually outputs two copies of the signal, one to be used for energy reconstruction and one for timing reconstruction. As inputs, the HPGe detector requires a low-voltage supply to power the preamplifier and the high-voltage supply to bias the detector. Additionally, the temperature of the crystal is monitored and a bias shutdown signal is produced. This signal will disable the high voltage in the event that the crystal warms up.

The high voltage for all HPGe detectors used is supplied by an ISEG NHQ 224M module. This NIM module is connected to the DAQ computer to allow remote biasing and monitoring of the detectors. Each detector has a built-in high-voltage filter to reduce electronic noise produced by the switching high-voltage power supply.

3.1.2 NaI annulus

Both experiments use NaI annuli produced by Saint-Gobain as an active veto surrounding the detectors. NaI (sodium iodide) is a scintillator commonly used for $\gamma$-ray detection. It has a much lower energy resolution than HPGe, around 5.5% FWHM
at 1 MeV. The advantages provided by NaI include that it is cheaper than HPGe, does not require cryogenic cooling, and NaI crystals can be grown much larger than is possible with germanium. $\gamma$ rays may escape the HPGe detectors after Compton scattering and depositing only part of their full energy. This background is reduced with a Compton suppression shield, here a NaI annulus. Furthermore, the active veto helps reduce natural radiation from the lead shielding, cosmic-ray induced events, and $\gamma$-ray cascades, where multiple $\gamma$ rays are produced from the same radioactive decay. The annulus vetoes $\approx 50\%$ of all events for both apparatuses. The reduction in background by the NaI annulus is shown in Fig. 3.1. NaI is especially desirable because it contains fewer radioactive impurities than other scintillators. The two-clover apparatus originally used a BGO (bismuth germanium oxide) active veto, but it was discarded in favor of NaI after discovering the large radioactive impurities present.

Like germanium detectors, charged radiation excites electrons in NaI from the valence band to the conduction band. The electron then de-excites by emission of a photon. The emitted light is of the correct wavelength to be reabsorbed by the scintillator, which decreases the light collection efficiency. This problem is remedied by doping the NaI with thallium impurities. This inserts a new energy level in the band-gap region between the valence and conduction band. The electron-hole pairs will migrate through the crystal lattice and decay through the impurity levels [65]. The scintillator is thus transparent to the produced light. PMTs are optically coupled to windows in the NaI shielding using optical grease. Scintillation light is converted into electrons at the PMT photocathode. The electrons then pass through a series of electrically biased dynodes which multiply the avalanche of electrons. The electrons are collected at the anode to produce a current that is recorded by the electronics [66].

For both experiments, the PMT high voltage is supplied by an ISEG NHQ 204M
NIM module. This module is connected to the DAQ computer and continuously monitored to ensure the PMTs remain biased.

3.2 Two-coaxial HPGe apparatus

This apparatus has previously measured the $2\nu\beta\beta$ decay of $^{100}$Mo and $^{150}$Nd to their first excited $0^+$ states [68, 69]. Furthermore, it was also used to set limits on the $0\nu E C E C$ decay of $^{112}$Sn [70]. The experiments conducted on $^{100}$Mo and $^{112}$Sn were done at ground level in the TUNL cave. During January of 2008, the apparatus was moved to KURF where the measurements on $^{150}$Nd were performed. In this work, it is used to investigate the $2\nu\beta\beta$ decay of $^{96}$Zr. This is the second experiment this apparatus has conducted while located at KURF.

The apparatus uses two coaxial Ortec HPGe detectors. The detectors are $p$-type crystals 88 mm in diameter and 50 mm in length. They are shown housed inside the NaI annulus and sandwiching the target of interest in Fig. 3.3. The attached dewars hold 30 liters of liquid nitrogen and are filled weekly. The NaI annulus measures 50 cm in length with an exterior diameter of 35.6 cm and interior diameter of 12.5 cm. Scintillation light from the NaI annulus is recorded by six 3” PMTs (three on each side). In addition to the NaI annulus, two plastic scintillator end caps are also used. These are placed at the ends of the annulus to provide coverage where the NaI annulus does not. Each plastic scintillator end cap is connected to two 2” PMTs. Although plastic is not an efficient $\gamma$-ray detector, the end caps help provide close to $4\pi$ veto coverage for the coaxial HPGe. This helps reduce backscattered $\gamma$ rays that would not be detected by the NaI annulus.

The detectors are surrounded by several layers of passive shielding. Closest to the NaI annulus is 3/4” oxygen-free high-conductivity (OFHC) copper, followed by 6” lead. The roof of the housing is supported by an additional 1/2” steel plate placed in between the copper and lead. The lead reduces $\gamma$ rays produced by natural
radioactivity in the mine rock. Copper is used for the innermost level of shielding as it contains far fewer radioactive isotopes and impurities than lead.

These detectors were specially designed with the preamplifier and high voltage filter located outside the lead shielding, next to the detector’s nitrogen dewar. Normally preamplifiers are located close to the crystal to obtain the best signal to noise ratio. As components in the preamplifier often contain large radioactive impurities, low background studies benefit from additional shielding between the detector and preamplifier.

3.2.1 Electronics

The electronics for this apparatus are already detailed in [71, 72]. A short summary of the important details is given here. All modules use the NIM standard, except where noted, and are linked via coaxial cables. Five signals are recorded for each generation of the master trigger: the pulse height of each HPGe, the timing between
the HPGe, and two copies of the veto timing.

Both detectors have their preamplifier output fed into a spectroscopic amplifier with a shaping time of 6 µs. The unipolar amplifier output is delayed and fed into an 11 bit EG&G Ortec ADC CAMAC module to record the pulse height and reconstruct the energy deposited in each detector. The bipolar amplifier output is fed into a timing Single Channel Analyzer (SCA) and discriminator to form a timing pulse for logic operations. The timing pulses for both detectors are summed and fed through a gate generator to form the master trigger for the ADC. The timing between the two detectors is measured with a TAC. The timing pulse from detector one is used as the start signal for the TAC, and the detector two timing pulse is delayed and used as the stop signal. The TAC signal inputs to the same ADC module.

The signals from the PMTs, both on the NaI annulus and the plastic end caps, are summed together by discriminators. This signal is joined with the master trigger signal in a logic ‘AND’ module and is fed into a TAC as the start signal. This reduces TAC dead time by only having the veto TAC fire for events in the HPGe detectors. A copy of the veto signal is delayed and used as a stop signal for this TAC. Two copies of this signal are fed into an ADC and record the timing between the master trigger and the veto trigger.

The CAMAC module and data acquisition are handled by CODA at TUNL (C@T), a version of Jefferson Lab’s CEBAF Online Data Acquisition software modified for TUNL [73]. The data are recorded on a Unix workstation and converted from the CEBAF common event format into ROOT TTrees using the program CODA2ROOT.

3.3 Two-clover HPGe apparatus

The two-clover apparatus was originally assembled in the TUNL cave (at ground level) during 2010. After extensive testing, characterization, and efficiency measurements, the apparatus and lead shielding was moved to KURF in 2012. The NaI
annulus was added (replacing a plastic annulus) in March of 2013. Since this time, the apparatus has been dedicated to counting E\(CEC\) of \(^{156}\text{Dy}\).

A schematic of the apparatus is shown in Fig. 3.4. Much like the two-coaxial apparatus, two clover HPGe detectors sandwich a target of interest and are surrounded by a NaI annulus. The liquid nitrogen dewars are shown at the end of the detectors and the preamplifiers are housed in between the crystal and dewar. The NaI annulus and lead shielding is also shown surrounding the detectors. Liquid nitrogen is filled daily by an automated fill system that was designed and built at TUNL. This system allows for remote monitoring and scheduling of fills. The details of two-clover apparatus and the differences from the two-coaxial apparatus are documented below.

### 3.3.1 Clover HPGe detectors

The primary innovation in this apparatus is the use of Canberra clover HPGe detectors. The efficiency of a \(\gamma\)-ray detector may be increased by simply increasing the size of the detector. Growing germanium crystals becomes difficult and expensive above a certain size, so composite germanium detectors were designed. These detectors feature multiple crystals sharing a common cryostat and achieve a larger active volume than any single crystal. The clover design features four coaxial-HPGe detectors (referred to as segments) arranged in a pattern reminiscent of a four-leaf clover, shown in Fig. 3.5. Each crystal is 50 mm in diameter and 80 mm in length before shaping. The detectors are extensively characterized in Ref. [74].

An important feature of the clover detector is that each segment acts as an individual detector. This means that two coincident \(\gamma\) rays may interact in two separate segments, allowing for complete energy reconstruction of both incident \(\gamma\) rays. Another scenario is when one \(\gamma\) ray Compton scatters, depositing its energy in multiple segments. This gives rise to a second mode of operation, known as addback, where the energy in each segment is summed together to reconstruct the energy of
3.3.2 Active shielding

The active shielding simply consists of the NaI annulus; no plastic scintillator end caps are used for the two-clover apparatus. The NaI annulus measures 17.164” long.
and has a 14.500” exterior and 6.000” interior diameter. It is housed in aluminum: 0.031” thick inside walls and 0.125” thick outside walls. Twelve Hamamatsu R329-03 2” PMTs, six on each side of the detector, monitor the scintillation light. The high voltage on each PMT was optimized to give the maximum gain without amplifying detector noise (1400-1600 V). The trigger threshold for this detector is $\approx 50$ keV.

The clover detectors were originally purchased with BGO (bismuth germanium oxide) scintillator shields designed for Compton suppression. These shields were quickly discovered to contain large amounts of radioactivity making them unsuitable for low background experiments. Strong $\gamma$-ray lines were seen from the decay of $^{214}$Bi, most likely due to contaminates of $^{226}$Ra in the BGO crystals and PMTs. The BGO shields were removed and temporarily replaced by two scintillating plastic annuli. The two-clover apparatus was housed inside these annuli while the apparatus was tested on the surface and before the NaI annulus arrived. The plastic annuli vetoed cosmic rays such as muons, but was not dense enough to work as a Compton suppression system. It was removed and replaced by the NaI annulus during the start of the $^{156}$Dy runs.
3.3.3 Passive shielding

The passive shielding primarily consists of a lead brick housing surrounding the detectors. The floors and side walls are shielded by 8” lead. A roof was constructed using a 123 × 91.5 × 2 cm copper plate, which support 6” lead. The roof is extended along the length of the annulus by one 41 × 91.5 × 2 cm iron plate on both sides of the copper. The iron plates supports 4” of lead. The lead was cleaned by acid etching at TUNL. After the lead was moved underground, the lead bricks were sanded to remove the outermost layer of oxidation from the lead. Figure 3.7 shows the lead before and after sanding.
3.3.4 Electronics

The data-acquisition system for the two-clover apparatus uses NIM electronics for all signal processing and logic operations. The signals are then read and digitized by VME electronics. With the exception of the ADC and TDC modules, all of the modules shown below are NIM standard. All modules and detectors are connected using coaxial cables, except for the ECL logic signals, which are connected using twisted pair cables.

For simplicity, the electronics are broken into three subsystems: the ADC circuit, the TDC circuit and trigger generation, and the NaI veto circuit. Each clover segment has its own preamplifier. One copy of the signal goes to the ADC circuit for energy reconstruction while the other copy is used in the TDC circuit for timing and generation of the master trigger.

The TDC circuit is shown in Fig. 3.8. The eight signals are first amplified by a timing-filter amplifier, then sent through a constant-fraction discriminator (CFD). If the pulse meets the CFD threshold (≈ 90 keV), a NIM logic pulse is produced that conserves the timing of the original pulse but now with uniform amplitude and width.

Figure 3.7: (Color online) Lead shielding for the two-clover apparatus. The pretreatment lead (left) and the cleaned lead forming the floor of the detector housing (right) are shown for comparison.
This filters out low energy noise and reduces the trigger rate of the detector. The pulses then travel through a gate generator. When the apparatus was at the surface, veto and coincidence cuts were implemented in hardware. The gate generator adjusts the pulse duration before any logic cuts are imposed. The trigger rate decreased after the apparatus was moved underground and the hardware cuts were removed in favor of performing the cuts in software, but the gate generator remained to provide the option of performing hardware cuts. Signals from all eight clover segments are then fed into an octal CFD. This module translates the NIM logic pulses into the ECL standard which are then measured by the the 12-bit TDC. The octal CFD also sums all eight signals to use for the trigger generation.

While the TDC and the ADC are processing pulses, they produce a busy signal. These busy signals are converted from ECL into NIM logic by a level translator and summed in a fan in/out module. The master trigger signal is created as the sum of the octal CFD given that neither the TDC or ADC is busy. The master trigger initiates the ADC and TDC to start recording. Pulses lost due to the busy signal are accounted for by the dead-time correction. Given the slow trigger rate (≈ 4.5 Hz) and the digitization time scale (80 µs) the dead-time correction is minuscule.

The ADC circuit, shown in Fig. 3.9, records the energy deposited in each detector.
Each crystal has its output amplified by a spectroscopic amplifier. The spectroscopic amplifier consists of a high-pass filter, amplifier, and low-pass filter. An integration time scale of 4 $\mu$s was used. This removes electronic noise and shapes the charge collection pulses from the detector into Gaussian pulses that can be read by the ADC. A 12-bit ADC converts the voltage amplitude of the pulse into a digital number between 0 and 4095 that is stored by the DAQ. The ADC records a value for each input during every trigger. This means that events below the CFD threshold are recorded as long as they are in coincidence with events that pass the CFD threshold and generate a master trigger signal.

Given the poor energy resolution of NaI, the energy spectrum would not be helpful in reconstructing events originating in the HPGe detectors. For this reason only the timing of the signal is recorded. The resulting cut on the data is then binary: either the veto fired, or it did not. The NaI circuit is shown in Fig. 3.10. The signals from all 12 PMTs are summed using a fan in/out module to form one signal for the annulus. One copy of this signal is amplified by a TFA while the other is left unamplified. Both signals are converted from NIM logic signals into ECL signals and fed into the same TDC module as the clover signals.

Data acquisition is handled by C@T, as discussed in 3.2.1. At every generation of the master trigger, each ADC and TDC channel are read out by a VME “read
out controller.” The data are sent via Ethernet cable to an event builder process on a Linux workstation, where they are written to a file in the CEBAF common event format. These files are then converted into a ROOT TTree using TRAP, the TUNL Real-time Analysis Package [73].

3.4 KURF

Both discussed experiments took place at KURF, an underground lab located in an active limestone mine in Ripplemead, Virginia. The lab is on the 14th level, with an overburden of 520 m of dolomite and limestone, providing 1450 m.w.e. (meters water equivalent) shielding from cosmic rays [75]. The muon flux at KURF and other underground labs is shown in Fig. 3.11. Although other labs provide a much lower muon flux, the overburden at KURF is sufficient for an experiment focused on the detection of \( \gamma \) rays. The reduction in the background spectra after moving the two-clover apparatus underground is shown in Fig. 3.12. Notable features include a lower continuum and a substantial reduction in the 511 keV annihilation peak. A noticeable decrease in the neutron background is shown by the disappearance of the 595.9 keV peak. This peak may either be caused by inelastic neutron scattering off \(^{74}\text{Ge}\) or thermal neutron capture on \(^{73}\text{Ge}\) in the detectors.

Both experiments are housed inside the conex container shown in Fig. 3.13. The container is outfitted with an air conditioning unit and HEPA air filters to remove dust from the mine air. A high-speed internet connection is present, allowing the DAQ and computers to be controlled remotely.

3.5 Samples

Samples enriched in the isotopes of interest were leased from ORNL. Both the zirconium and dysprosium samples are oxide powders. The samples were isotopically enriched in the ORNL calutrons. These were mass spectrometers invented by Ernest
Figure 3.11: (Color online) The muon flux at KURF. Other underground laboratories are shown for comparison. Taken from [76].

Figure 3.12: (Color online) The spectra from the two-clover apparatus at TUNL (ground level) and at KURF.
Lawrence and used a cyclotron’s magnetic field to separate different isotopes [77]. The calutrons were originally used to enrich uranium for the Manhattan project. These samples were created long after ORNL discontinued enriching uranium, but they still contain uranium and thorium contaminates from the calutron process. Given the high level of radiopurity in HPGe detectors, shielding, and environment, the sample can then be the largest source of background for these experiments.

3.5.1 96Zr sample

Two ZrO₂ samples of different enrichment were obtained from ORNL. Their isotopic enrichment is summarized in Table 3.1. Since the efficiency of the two coaxial detector apparatus is the highest at the center of the detectors faces, the more enriched sample (#1) was concentrated at the center of the apparatus. This sample has a mass of 7.2835 g, resulting in a mass of 4.987 g 96Zr. The sample was packaged in an acrylic
Figure 3.14: (Color online) Inside of the conex container. The housing for the two-clover apparatus is shown on the left, the electronics for the two clover apparatus are on the right. The dewars and lead housing for the two-coaxial apparatus may be seen in the background.

Table 3.1: Isotopic enrichment for ZrO$_2$ samples. For comparison, the natural abundance of $^{96}$Zr is 2.8%.

<table>
<thead>
<tr>
<th>Isotope Zr</th>
<th>Sample 1</th>
<th>Sample 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>90</td>
<td>3.95 %</td>
<td>21.21 %</td>
</tr>
<tr>
<td>91</td>
<td>0.88 %</td>
<td>2.30 %</td>
</tr>
<tr>
<td>92</td>
<td>1.39 %</td>
<td>16.25 %</td>
</tr>
<tr>
<td>94</td>
<td>2.38 %</td>
<td>5.07 %</td>
</tr>
<tr>
<td>96</td>
<td>91.39 %</td>
<td>64.18 %</td>
</tr>
</tbody>
</table>

disk with 1.4 mm thick walls, 31.75 mm outer diameter, and 10.22 mm height. The height of the sample mimics the dimensions of previous measurements [68].

Sample #2 has a mass of 26.9685 g oxide, 12.927 g $^{96}$Zr, and is housed in an annular acrylic container designed to encompass the more enriched sample. This container has a 69.95 mm outer diameter, 10.07 mm height, 1.31 mm radial walls, and 1.57 mm thick longitudinal walls. The two samples together total 17.914 g $^{96}$Zr and were centered on the face of the coaxial detectors.
Table 3.2: Isotopic enrichment for Dy$_2$O$_3$ samples. For comparison, the natural abundance of $^{156}$Dy is 0.056%.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Sample 1</th>
<th>Sample 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{156}$Dy</td>
<td>21.59%</td>
<td>20.9%</td>
</tr>
<tr>
<td>$^{158}$Dy</td>
<td>0.36%</td>
<td>0.58%</td>
</tr>
<tr>
<td>$^{160}$Dy</td>
<td>3.10%</td>
<td>3.43%</td>
</tr>
<tr>
<td>$^{161}$Dy</td>
<td>19.29%</td>
<td>19.7%</td>
</tr>
<tr>
<td>$^{162}$Dy</td>
<td>20.69%</td>
<td>22.2%</td>
</tr>
<tr>
<td>$^{163}$Dy</td>
<td>17.70%</td>
<td>16.9%</td>
</tr>
<tr>
<td>$^{164}$Dy</td>
<td>17.26%</td>
<td>16.3%</td>
</tr>
</tbody>
</table>

3.5.2 $^{156}$Dy sample

Once again, two samples of varying enrichment were obtained from ORNL. The enrichment of the samples is detailed in Table 3.2. The samples were placed in polycarbonate bags. Sample #1 is 803.4 mg oxide, or 150.95 mg $^{156}$Dy. It was placed inside a 0.089 mm thick bag with exterior dimensions $3.8 \times 3.3 \times 0.14$ cm. Sample #2 is the smaller sample: 344.3 mg oxide, 62.62 mg $^{156}$Dy. This sample was placed in a $3.5 \times 2.8 \times 0.165$ cm bag. Sample #2 was placed in multiple bags, such that the effective bag thickness is 0.762 mm. The samples were placed on top of each other inside a 0.089 mm thick bag, resulting in a total thickness of 0.375 cm and a total mass of 213.57 mg $^{156}$Dy. The samples were centered on the face of the clover detectors.

3.6 Previous measurements

Previous attempts to measure the $\beta\beta$ decay of $^{96}$Zr and ECEC decay of $^{156}$Dy to excited states used a single HPGe detector. If a single detection experiment has efficiency $\epsilon$, a coincidence experiment with two identical detectors would have efficiency $O(\epsilon^2)$. The trade off for the reduced efficiency of a coincidence measurement is a decrease in background, resulting in decreased statistical uncertainties and a much higher signal to background ratio.
Other experiments have used multiple HPGe detectors in coincidence to search for \(\beta\beta\) decay to an excited state. Two HPGe detectors arranged in a similar configuration were used to search for the \(2\nu\beta\beta\) decay of \(^{102}\text{Pd}\) and \(^{110}\text{Pd}\) to their \(0^+_1\) state in Ref. [78]. The DAQ system for this experiment was limited, however, and the coincidence technique was not employed. Four HPGe detectors were used in Ref. [79] to set limits on the \(\beta\beta\) decay of \(^{150}\text{Nd}\) and \(^{76}\text{Ge}\) to their \(0^+_1\) states. A similar apparatus using four HPGe detectors was used to measure the \(\beta\beta\)-decay half-life of \(^{100}\text{Mo}\) to its \(0^+_1\) state [80]. Although not applied to second order weak decays or used in a low background setting, two clover HPGe detectors have previously been used in close geometry [81]. The improvement in the signal to background performance by using the coincidence technique with a two-clover apparatus is shown in Ref. [81].

3.6.1 On \(^{96}\text{Zr}\)

The current best limits use a 430 cm\(^3\) well-type HPGe detector and are reported in Ref. [82]. Well type detectors invert the coaxial design such that the samples may be placed inside the central hole. Well detectors thus cover a large solid angle and have high efficiencies, but the sample size is restricted to that of the hole. Well-type detectors also typically suffer from poorer resolution and issues resulting from accidental summing of coincident \(\gamma\) rays.

Reference [82] used a 18.74 g sample of \(\text{ZrO}_2\) enriched to 57.3\% in \(^{96}\text{Zr}\), resulting in 8.0 g \(^{96}\text{Zr}\). The experiment was carried out in the Laboratoire Souterrain de Modane in France, 4800 m.w.e. The sample was counted for 464.4 hours. The same sample was used in another experiment and the results were presented in Ref. [83, 84]. This time a 314 cm\(^3\) coaxial HPGe detector 81 mm in height and 71.6 mm in diameter was used. The detector was housed at LNGS in Italy, 3600 m.w.e. 2503 hours of data were collected. The results of both these experiments are summarized in Table 3.3.
Table 3.3: Limits on $\beta\beta$ decay of $^{96}$Zr to excited states at CL = 90%.

<table>
<thead>
<tr>
<th>Transition</th>
<th>$E_{exc}$ keV</th>
<th>Lim $T_{1/2}$ yr</th>
</tr>
</thead>
<tbody>
<tr>
<td>$0^+ \rightarrow 2^+_1$</td>
<td>778.2</td>
<td>$4.1 \times 10^{19}$</td>
</tr>
<tr>
<td>$0^+ \rightarrow 0^+_1$</td>
<td>1147.9</td>
<td>$3.3 \times 10^{19}$</td>
</tr>
<tr>
<td>$0^+ \rightarrow 2^+_2$</td>
<td>1497.7</td>
<td>$2.4 \times 10^{19}$</td>
</tr>
<tr>
<td>$0^+ \rightarrow 2^+_3$</td>
<td>1625.7</td>
<td>$3.1 \times 10^{19}$</td>
</tr>
<tr>
<td>$\beta$ decay</td>
<td>$3.8 \times 10^{19}$</td>
<td>$2.6 \times 10^{19}$</td>
</tr>
</tbody>
</table>

Limits on the single-$\beta$ decay of $^{96}$Zr are shown as well. These limits were found by searching for the 778.2 keV $\gamma$ ray resulting from the $2^+ \rightarrow 0^+$ part of the cascade (see Fig. 2.1). Additional losses in efficiency are taken due to accidental summing of coincident $\gamma$ rays from the cascade. It is important to note that observing single $\gamma$ rays inherently limits observation of $\beta$ and $\beta\beta$ decay as either process could produce a 778.2 keV $\gamma$. By measuring multiple $\gamma$ rays in coincidence, the two decays can be distinguished with high efficiency. The present experiment further improves on these measurements by having a larger sample mass by a factor of 2.2.

3.6.2 On $^{156}$Dy

The leading measurements on $ECEC$ of $^{156}$Dy were performed at LNGS using a single 244 cm$^3$ coaxial HPGe detector and are reported in [85]. A 322 g sample of $^{nat}$Dy$_2$O$_3$ was used. This large sample was of natural abundance, 0.056%, and only amounts to 157 mg of $^{156}$Dy. The resulting limits after 2512 hours of counting are shown in Table 3.4.

The current work improves by having a 36% larger sample when both samples are utilized. Furthermore, when using a large 322 g sample, numerous corrections must be made to the efficiency. Firstly, there will be a sizable self attenuation by the sample. With the enriched samples used in the work, this effect is close to negligible. Secondly, numerous simulations are necessary to determine the effect of the source’s spatial extent on the efficiency. Additionally, the efficiency for a large sample will be
Table 3.4: Limits on \( ECEC \) decay of \(^{156}\text{Dy}\) to excited states at CL = 90%.

<table>
<thead>
<tr>
<th>Q (keV)</th>
<th>( E_{Exc} ) (keV)</th>
<th>( J^\pi )</th>
<th>e(^-) orbitals</th>
<th>Lim T(_{1/2}) [85] yr</th>
</tr>
</thead>
<tbody>
<tr>
<td>1905.5</td>
<td>1914.8</td>
<td>2(^+)</td>
<td>2K</td>
<td>( 1.1 \times 10^{16} )</td>
</tr>
<tr>
<td>1947.3</td>
<td>1946.4</td>
<td>1(^-)</td>
<td>KL(_1)</td>
<td>( 9.6 \times 10^{15} )</td>
</tr>
<tr>
<td>1947.3</td>
<td>1952.4</td>
<td>0(^-)</td>
<td>KL(_1)</td>
<td>( 2.6 \times 10^{16} )</td>
</tr>
<tr>
<td>1989.2</td>
<td>1988.5</td>
<td>0(^+)</td>
<td>2L(_1)</td>
<td>( 1.9 \times 10^{16} )</td>
</tr>
<tr>
<td>1991.5</td>
<td>2003.7</td>
<td>2(^+)</td>
<td>2L(_3)</td>
<td>( 3.0 \times 10^{14} )</td>
</tr>
</tbody>
</table>

much lower since a single detector covers a smaller effective solid angle.

Another concern in [85] is the multiplicity of the \( \gamma \) cascade. Most of the excited states from resonant \( ECEC \) will cascade emitting a multitude of \( \gamma \) rays. The angle between \( \gamma \) rays is dictated by a probability density function \( W(\theta) \). In order to observe the full excitation energy \( E_{Exc} \), all of these \( \gamma \) rays must be emitted in approximately the same direction (towards the detector) and deposit all of their energy in the detector. The present work covers closer to \( 4\pi \) by surrounding the sample with two clover detectors. This allows for detection of back to back \( \gamma \) rays in addition to two forward going \( \gamma \) rays, effectively covering a larger portion of \( W(\theta) \).
A number of analyses were conducted using the two-coaxial HPGe detector apparatus previously discussed. The apparatus was used to search for the double-\(\beta\) decay to excited states and the single-\(\beta\) decay of \(^{96}\text{Zr}\). This chapter will discuss the cuts applied to the data, efficiency calculations, and backgrounds for all of these experiments. The entire analysis was performed using CERN’s ROOT framework.

4.1 Data collection and quality control

Data collection with the \(^{96}\text{Zr}\) sample took place over 1134.9 days. The detector live time over this period was 702.1 days (63.7%), distributed as shown in Fig. 4.1. There are two large down times during this period. The first occurred at the beginning of data collection and the second at day 947. After substantial troubleshooting, both were determined to be the result of deterioration of the vacuum in detector one. In both cases, the detector was sent back to Ortec for vacuum repair work.

Given the large time scale of the present experiment, the long term stability of the detectors was investigated by monitoring the detector’s count rate, resolution,
Figure 4.1: (Color online) The live time of the detectors with the $^{96}$Zr sample in place. Up-time is colored red. The two large outages at the beginning of data collection and at day 947 correspond to when detector 1 was sent in for vacuum repairs.

Figure 4.2: (Color online) The FWHM of each detector as a function of time. The short runs and low count rate contributes to the large statistical uncertainties. The resolution increase in detector 1 at 900 days prompted the detector to be sent in for vacuum repairs.

and efficiency. The resolution as a function of time is shown in Fig. 4.2. The FWHM of both detectors is evaluated for each run by fitting the 583.2 keV peak (a natural background $\gamma$ ray from $^{208}$Tl’s $\beta^-$ decay to $^{208}$Pb) to a Gaussian. With the low count rate of the experiment, large statistical uncertainties are present in the measurement of the detector FWHM for some of the shorter runs. The increases in the FWHM of detector one are what prompted the detector to be sent in for vacuum repairs, as has already been mentioned.

Detector stability was also investigated through the integration of natural background peaks. An intense 1460.8 keV $\gamma$ ray is produced by the electron-capture decay of $^{40}$K. $^{40}$K is present in the limestone rock and has a long half-life ($10^9$ yr), resulting
Figure 4.3: (Color online) The yield in the 1460.8 keV $\gamma$ ray following the electron-capture decay of $^{40}$K.

in a constant concentration over the course of the experiment. A constant yield in the 1460.8 keV peak can be used as a verification of a constant detector efficiency. Figure 4.3 shows the effect on a run by run basis. The outliers are all short runs where the uncertainties are dominated by statistics.

The same analysis is also done for the 609.3 keV $\gamma$ ray following the $\beta$ decay of $^{214}$Bi. $^{214}$Bi is produced in the $^{238}$U decay chain, following the production of radon. As a noble gas with a 3.8 day half-life, radon can travel from its source to plate out on detector components and shielding. The Kimballton lab level stays at the same temperature year round, ignoring the large seasonal temperature changes at the mine entrance. This causes better air circulation in the winter months, resulting in lower radon levels in KURF. This effect is seen very clearly in Fig. 4.4. Indeed, the two detectors are found to have periods of $381 \pm 2$ and $373 \pm 2$ days, respectively. Fixing the period at 365.25 days produces an annual maxima on July 14th and 13th ($\pm 1$) day for the two detectors. This is of some importance given the recent interest in searching for annual modulations produced by hypothetical dark matter WIMPs.
Figure 4.4: (Color online) The yield in the 609.3 keV $\gamma$ ray following the $\beta$ decay of $^{214}$Bi.

4.2 Data processing

One of the strengths of the presented experiment is the simplicity of the analysis, with minimal data cuts and statistical fits. Here all elements of data processing are discussed. Data are divided into runs, which are manually started and stopped approximately every 4 days, although in some cases runs will vary between 1 and 6 days in length.

First, a histogram of the ADC readout is calibrated to energy in keV. This is done individually for each run. Four background peaks (238.6, 511.0, 1460.8, and 1764.5 keV) are fitted with a Gaussian and the centroid channel is plotted as a function of energy. A linear function is then used to convert from channel to energy. The amplifiers used in the present experiment are sensitive to temperature changes that might induce gain shifts into the data. Each individual run is examined for gain shifts that degrade detector resolution, and runs exhibiting such behavior are excluded from the analysis. A further examination of detector stability is found by investigating the calibration as a function of time. Figure 4.5 shows the ADC channel corresponding to a calibrated energy of 500 keV as a function of time for both detectors. The shifts are plotted as a deviation from average. Even though
Figure 4.5: (Color online) The long term stability of the two coaxial detectors is illustrated. Deviations from the average are plotted as a function of time for a calibration of a 500 keV $\gamma$ ray. Detectors one and two are shown by the black and red markers, respectively.

Table 4.1: Efficiency and experimental background rates in the region of interest. The efficiencies are taken from unbenchmarked GEANT4 simulations and only meant for comparison purposes.

<table>
<thead>
<tr>
<th>Region of interest</th>
<th>Efficiency</th>
<th>Background rate</th>
</tr>
</thead>
<tbody>
<tr>
<td>369 keV</td>
<td>17.0 %</td>
<td>21 cts/(keV day)</td>
</tr>
<tr>
<td>778 keV</td>
<td>9.8 %</td>
<td>7.3 cts/(keV day)</td>
</tr>
<tr>
<td>1148 keV</td>
<td>2.47 %</td>
<td>1.5 cts/(keV day)</td>
</tr>
<tr>
<td>369+778 keV</td>
<td>2.48 %</td>
<td>&lt;0.002 cts/(keV day)</td>
</tr>
</tbody>
</table>

each run is individually calibrated, we still observe excellent stability of $<0.5\%$ for the majority of runs without calibration. All runs passing the above data quality test are analyzed using the same procedure.

The following cuts will be chosen in order to maximize the signal to background of the apparatus. A quick investigation of the different regions of interest for $^{96}\text{Zr}$'s $\beta\beta$ decay to the first excited $0^+$ state is shown in Table 4.1. The first two rows represent detection of a single $\gamma$ ray from the decay, which has the highest efficiency as well as the highest background rate. Row three details the detection of both $\gamma$ rays in the same detector. The final row shows a comparable efficiency is obtained using the two detectors in coincidence, but the background rate is reduced by several orders of magnitude.
Figure 4.6: (Color online) The TAC spectra for the NaI+plastic veto on the two-coaxial HPGe apparatus.

The first cut applied to the data is an anticoincidence cut on the TAC signal for the NaI and plastic vetoes. The benefits of the veto were discussed in detail in the previous chapter. A typical TAC spectra is shown in Fig. 4.6 with the cut values illustrated by the red lines. The first and last hundred channels are not vetoed due to nonlinearities in the ADC response at high and low channels.

In order to impose the coincidence criteria discussed above, a second cut is applied on the coincidence TAC between the two detectors. This is shown in Fig. 4.7 with the cut values again illustrated by red lines. The results of this cut and the veto cut may be seen in Fig. 3.1. A 2D histogram of the coincidences is shown in Fig. 4.8. In this plot, pairs of coincident $\gamma$ rays create intense points. Given that the detectors are identical and the geometry symmetric, the plot is symmetric as well. Every coincidence $(\gamma_1, \gamma_2)$ will have a corresponding point $(\gamma_2, \gamma_1)$. When one of these $\gamma$ rays Compton scatters in a detector, depositing only part of its energy, the vertical and horizontal lines are produced and terminate at the coincident point. The diagonal lines are caused by intense single $\gamma$ rays that Compton scatter from one detector into the other. The energy is shared between the two detectors such that the sum of the energy in each detector is the energy of the incident $\gamma$ ray.

The final cuts are on the energy deposited in each detector. These cuts are a
Figure 4.7: (Color online) The coincidence TAC spectra for the two-coaxial HPGe apparatus.

function of the detector’s energy resolution. The FWHM is measured for each detector using the complete data set over the entire run period. The detector resolution is measured for six different peaks and extrapolated as a linear function of incident $\gamma$-ray energy, as shown in Fig. 4.9. Energy cuts are performed as $\pm N\sigma$, where $\sigma$ is the width of the Gaussian signal.

Figure 4.8: (Color online) The 2D coincidence spectra for the two-coaxial HPGe apparatus showing the energy deposited in each detector.
Figure 4.9: (Color online) The energy dependent resolution of both detectors over the entirety of runs with the $^{96}$Zr sample in place. Note $FWHM \approx 2.355\sigma$.

4.3 $\beta\beta$-decay results

After applying the energy cuts in each detector, we are left with two histograms: one containing all events coincident with a $369.7 \pm N\sigma$ keV $\gamma$ ray in one detector and one containing all events in coincidence with a $778.2 \pm N\sigma$ keV $\gamma$ ray in one detector. These histograms are shown in Fig. 4.10 for a $N = 3$ cut width.

All background lines appearing in these histograms have been identified. The most prominent line is the $778.9 + 344.3$ keV coincidence from $^{152}$Gd. A number of intense lines Compton scatter, sharing their energy between the two detectors. In the histogram of events in coincidence with $369.7$ keV, this accounts for the $213.5$ keV peak from Compton scatters of $^{208}$Tl’s $583.2$ keV $\gamma$ ray, $239.6$ keV from $^{214}$Bi’s $609.3$ keV, $541.5$ keV from $^{228}$Ac’s $911.2$ keV, $631.3$ keV from $^{234m}$Pa’s $1001.0$ keV, and $1091.1$ keV from $^{40}$K’s $1460.8$ keV. For events in coincidence with $778.2$ keV, this causes the $682.6$ keV peak from $^{40}$K’s $1460.8$ keV peak, $983.3$ keV from $^{214}$Po’s $1764.5$ keV, $1069.2$ keV from $^{214}$Bi’s $1847.4$ keV, and $1836.5$ keV from $^{208}$Tl’s $2614.5$ keV. Other intense lines present in both spectra are from a naturally occurring $\gamma$-ray coincidence where one $\gamma$ ray Compton scatters into the coincidence of interest. This includes the $511$ keV annihilation peak, $569.8 + 1063.6$ keV from inelastic neutron
Figure 4.10: (Color online) The top histogram shows events in coincidence with $369.7 \pm 3\sigma$ and the bottom histogram shows events in coincidence with $778.2 \pm 3\sigma$. The background extrapolation regions for the $\beta\beta$ decay of $^{96}\text{Zr}$ to the $0^+_1$ state are shown by the red shaded regions. The blue shaded region shows the decay’s ROI. Scattering on lead, and $583.2 \pm 2614.5$ keV from $^{208}\text{Tl}$.

An observation of $\beta\beta$ decay to the $0^+_1$ state would produce a peak at 778.2 keV in the histogram of events given a 369.7 keV event and vice versa. Histograms focused around this region of interest (ROI) are shown in Fig. 4.11 for both $N = 2$ and $N = 3$ cut width. The background level is extrapolated as a flat line around the ROI. The background extrapolation region was taken as the largest peak-free region surrounding the ROI, as shown in Fig 4.10. It should be noted that this region will contain weaker peaks from natural background transitions (mostly in coincidence with a Compton-scattered $\gamma$ ray), and as such may slightly overestimate the background. This method of background estimation is chosen because it produces conservative limits. One prominent background line present in these ROI histograms is 785.6 keV. This line is from the $\beta^-$ decay of $^{212}\text{Bi}$ and is in coincidence with a
Figure 4.11: (Color online) The $\beta\beta$ decay to the first excited $0^+$ state region of interest with the $^{96}$Zr sample in place (702.1 days). (a) and (b) use the $\pm 2\sigma$ energy cut, while the (c) and (d) use the $\pm 3\sigma$ cut. (a) and (c) are in coincidence with 369.8 keV and (b) and (d) are in coincidence with 778.2 keV. Events matching the cuts are highlighted in red. The minimum detectable signal above background at the 90% confidence level is shown by the blue curve.

727.3 keV $\gamma$ ray that Compton scatters into the 369.8 keV ROI.

As no peak is seen above background, a limit is set for the decays. The minimum detectable Poisson signal above a known Poisson background is given by Feldman-Cousins statistics [86]. The 90% confidence level is used. The blue curve in Fig. 4.11 shows the expected flat background and the expected Gaussian signal. The most statistically sensitive region was found to be the $N = 3$ energy cut for events in coincidence with 778.2 keV, searching for a peak at 369.7 keV. This region was used to set limits on the decay.

The same procedure is performed for $\beta\beta$ decay to higher $0^+$ and $2^+$ excited states. Figure 4.12 shows the higher excited states of $^{96}$Mo following $\beta\beta$ decay. The results for these decays are shown in Fig. 4.13. Note that for the 2622 keV state, the intense
Figure 4.12: Level diagram for $\beta\beta$ decay in $^{96}$Zr to different excited states of $^{96}$Mo.

2614.6 keV background $\gamma$ ray from $^{208}$Tl will Compton scatter, leaving a peak just shy of the region of interest. Folded with detector resolution, this is the likely cause of the one event passing the cuts for the 2622 keV decay. The 2614 keV background was fit using the collected data and an estimated 1.20 events will leak into the signal region. These 1.2 counts were added to the expected background level shown by the blue curve in Fig. 4.13. All the other regions agree with the expected background to within $\approx 1\sigma$.

An independent comparison of the background may be done by looking at the same ROI while the same apparatus was investigating different samples. In addition to the $^{96}$Zr sample, the detectors counted 180.76 days with no sample in place and 642.81 days with a $^{150}$Nd sample in place. As a significant fraction of the background is attributable to the sample, background rates extracted from these measurements were not used in calculating the limits presented for $^{96}$Zr. Furthermore, having a sample in between the detectors attenuates the radioactive background in one detector from being seen by the other, thus changing the background inherent to the apparatus. It should be noted that the $^{150}$Nd sample was also produced in the ORNL calutrons and is expected to have similar radioactive impurities. The results of this analysis is shown in Table 4.2. The ROI and background estimation regions used were identical to those of the $^{96}$Zr analysis. This was done to make the numbers directly comparable, although an optimized background estimation would provide
Figure 4.13: The region of interest for the $\beta\beta$ decay of $^{96}$Zr to higher excited states. All histograms use the $N = 2$ energy cut. Events passing the cut criteria are highlighted red, with the expected signal at the given 90% confidence level shown by the blue curve. (a) and (b) show the $0^+ 778.2 + 551.8$ keV coincidence, (c) and (d) show the $2^+ 778.2 + 719.6$ keV coincidence, (e) and (f) show the $2^+ 778.2 + 847.8$ keV coincidence, and (g) and (h) show the $0^+ 778.2 + 1844.3$ keV coincidence.
Table 4.2: The same analysis on the $^{96}$Zr ROI was done for the background and $^{150}$Nd runs performed while the apparatus was underground. The $^{96}$Zr run lasted 702.1 days, the background run lasted 180.76 days, and the $^{150}$Nd run 642.81 days. For each run, the number of counts in each ROI $N_{obs}$ and the expected background $N_{bkgd}$ are given. See text for further discussion.

<table>
<thead>
<tr>
<th>$J^\pi$</th>
<th>$^{96}$Zr empty</th>
<th>$^{150}$Nd</th>
</tr>
</thead>
<tbody>
<tr>
<td>0$^+_1$</td>
<td>4 5.34 0 4.42</td>
<td>5 7.53</td>
</tr>
<tr>
<td>0$^+_2$</td>
<td>3 1.50 0 1.04</td>
<td>4 1.95</td>
</tr>
<tr>
<td>2$^+_2$</td>
<td>2 0.97 0 0.99</td>
<td>1 1.56</td>
</tr>
<tr>
<td>2$^+_3$</td>
<td>2 0.87 3 0.71 3</td>
<td>1 0.67</td>
</tr>
<tr>
<td>0$^+_3$</td>
<td>1 1.27 0 0.14</td>
<td>3 1.54</td>
</tr>
</tbody>
</table>

more accurate results. The width of the ROI, $\sigma$, was varied to reflect the detector conditions for each separate source. Detector one experienced vacuum degradation issues during the background runs, resulting in an increased $\sigma$. This caused some neighboring background lines to bleed into the ROI for the 2$^+_3$ state.

4.4 $\beta\beta$-decay efficiency calculation

The coincidence efficiency of the apparatus was measured using a $^{102}$Rh source. This source was chosen because it exhibits the $0^+ \rightarrow 2^+ \rightarrow 0^+$ decay scheme of interest for $\beta\beta$ decay to excited states. Furthermore, it also exhibits a $2^+ \rightarrow 2^+ \rightarrow 0^+$ decay mode. The complete decay scheme is shown in Fig. 4.14. $^{102}$Rh has a half-life of $207.3 \pm 1.7$ days, making it convenient for efficiency measurements. The $^{102}$Rh source was produced using the $^{102}$Ru(p,n)$^{102}$Rh reaction with 5 MeV protons from the TUNL tandem accelerator. The activity of the source was determined by comparing the 475.1 keV yield to the 661.6 keV yield of a calibrated $^{137}$Cs source.

The efficiency was measured as a function of radius for eight points between $0 \leq r \leq 4.5$ cm. Before showing the results of this measurement, a number of corrections must be made to the $^{102}$Rh measurement in order to be applicable to our measurement on $^{96}$Zr.
4.4.1 Energy dependence

HPGe detectors have a strong efficiency dependence on the energy of the incident $\gamma$ ray. This effect was measured using a number of point sources: $^{133}$Ba, $^{60}$Co, $^{137}$Cs, $^{152}$Eu, $^{22}$Na, and $^{54}$Mn. As the activity of each of these sources is only known to 3%, only the $^{152}$Eu source, which produces a number of $\gamma$ ray lines, was used for the calculations shown here. The efficiency as a function of energy is shown in Fig. 4.15. This dependence is confirmed by GEANT4 simulations of the detectors, also shown in the same plot. The GEANT4 model of the HPGe detector may be seen in Fig. 4.16.

The data are fit to a function of the form

$$\epsilon_\gamma(E) = \left( \frac{E}{a} \right)^b.$$  \hspace{1cm} (4.1)

The difference in efficiency is calculated as the ratio between the 369+778 keV and the 468+475 keV coincidences:

$$\frac{\epsilon_{\gamma\gamma}(369, 778)}{\epsilon_{\gamma\gamma}(468, 475)} = \frac{\epsilon_{\gamma1}(369)\epsilon_{\gamma2}(778) + \epsilon_{\gamma1}(778)\epsilon_{\gamma2}(369)}{\epsilon_{\gamma1}(468)\epsilon_{\gamma2}(475) + \epsilon_{\gamma1}(475)\epsilon_{\gamma2}(468)} = 0.827 \pm 0.004,$$  \hspace{1cm} (4.2)

where the numerical subscripts refer to detector 1 and 2. As we are only taking the ratio of two efficiencies, the systematic uncertainty of the $^{152}$Eu source’s calibration cancels out leaving a correction with a small relative uncertainty. The same procedure is performed for all coincidences of interest and is summarized in Table 4.3.
Figure 4.15: (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.

Figure 4.16: (Color online) A coaxial HPGe as modeled in GEANT4. The crystal is shown in green, the magnesium end cap window in red, and the aluminum shell in blue.

4.4.2 Target attenuation

The γ rays emitted by the target will be attenuated by the target itself. This effect may be accurately calculated by GEANT4 simulations. The efficiency measurements were taken when the apparatus was examining the $2\nu\beta\beta$ decay of $^{100}$Mo. In order to accurately reproduce the attenuation of the $^{100}$Mo sample, 5 mm of nat Mo metal were placed on both sides of the $^{102}$Rh point source. A GEANT4 simulation was run with the detector geometry, point source, and metal disk accurately reproduced. The same simulation was run without the nat Mo metal disk. The ratio of full energy coincidence counts in the $^{102}$Rh region of interest with and without the metal disk
Figure 4.17: (Color online) A side and top view of the $^{96}$Zr sample as modeled in GEANT4. The dimensions of the two samples and acrylic holder are given in Sec. 3.5.1.

Figure 4.18: (Color online) The two-coaxial HPGe apparatus as modeled in GEANT4. The $^{96}$Zr sample is shown in between the detectors.

gives an attenuation ratio. Although the attenuation ratio varies as a function of position, it is $\approx 3.27$. Each data point in Fig. 4.20 has been corrected by this method to accurately reflect the efficiency with no target attenuation.

The same procedure was performed in order to calculate the attenuation in the $^{96}$Zr target. The GEANT4 reconstruction of the $^{96}$Zr samples is shown in Fig. 4.17 and the target in between the two coaxial detectors is shown in Fig. 4.18. The attenuation must be calculated for each $\gamma$-ray pair since the attenuation is energy dependent. Results are summarized in Table 4.3.
Table 4.3: Efficiency correction ratios for different coincidences in $^{96}$Mo. The attenuation ratio of the $^{96}$Zr sample and the energy dependence ratio of the detectors are both given.

<table>
<thead>
<tr>
<th>$J^\pi$</th>
<th>$\gamma_1$ [keV]</th>
<th>$\gamma_2$ [keV]</th>
<th>Sample attenuation</th>
<th>$\epsilon(\gamma_1, \gamma_2)/\epsilon(468, 475)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$0^+_1$</td>
<td>369.7</td>
<td>778.2</td>
<td>0.851 ± 0.001</td>
<td>0.827 ± 0.003</td>
</tr>
<tr>
<td>$0^+_2$</td>
<td>551.8</td>
<td>778.2</td>
<td>0.868 ± 0.001</td>
<td>0.615 ± 0.006</td>
</tr>
<tr>
<td>$2^+_2$</td>
<td>719.6</td>
<td>778.2</td>
<td>0.875 ± 0.001</td>
<td>0.506 ± 0.007</td>
</tr>
<tr>
<td>$2^+_3$</td>
<td>847.7</td>
<td>778.2</td>
<td>0.881 ± 0.002</td>
<td>0.448 ± 0.007</td>
</tr>
<tr>
<td>$0^+_3$</td>
<td>1844.3</td>
<td>778.2</td>
<td>0.897 ± 0.002</td>
<td>0.252 ± 0.007</td>
</tr>
</tbody>
</table>

4.4.3 Detector separation

The separation distance between the two detectors can have a noticeable effect on the efficiency, as it is strongly correlated with the solid angle covered by the detectors. The efficiency measurements were done with an 11 mm separation distance. In the $^{96}$Zr experiment, the detectors are separated by the thickness of the sample: 10.22 mm. This effect was simulated using GEANT4. This results in a change of efficiency of

$$\frac{\epsilon(d = 10.22)}{\epsilon(d = 11.0)} = 1.022 \pm 0.0014. \quad (4.3)$$

This result may also be calculated theoretically. For a detector with radius $a$, source with radius $s$, along a common axis with separation distance $d$, the effective solid angle viewed by the detector is

$$\Omega = \frac{4\pi a}{s} \int_0^\infty \frac{exp(-kd)J_1(ak)J_1(sk)}{k} \, dk, \quad (4.4)$$

where $J_1$ are the Bessel functions of the first kind. Use of this formula produces results that agree well with the simulation. The simulation results were used as they contain corrections from higher order effects, such as the bulletization of the HPGe detector, and as such should be more accurate.
4.4.4 Z dependence of the efficiency

A GEANT4 simulation was conducted to determine the effect of the sample’s spatial extent along the Z-axis. The efficiency data are taken with a point source located at $Z = 0$ mm. Of course, our sample has a thickness of approximately 7 mm, so the actual decays will be uniformly distributed from $-3.5 < Z < 3.5$ mm. The GEANT4 simulation was performed for both of these conditions. This resulted in a ratio of efficiencies of

$$\frac{\epsilon(-3.5 < Z < 3.5)}{\epsilon(Z = 0)} = 0.9998 \pm 0.0014. \quad (4.5)$$

As no effect was seen at the level of statistical significance, this ratio was assumed to be unity, although the statistical uncertainty is added into the final error budget.

4.4.5 Angular correlation of coincident $\gamma$s

In $\gamma$ decay, the angular momentum $l$ of each state dictates the type of electromagnetic multipole radiation emitted during the decay. Multipole radiation has a determined angular distribution [87], although this is not evident for $\gamma$ decay at room temperatures. Successive decays will, however, lead to uneven population of the $m$ states, and distinct angular distributions between the two coincident $\gamma$ rays will be observed. These angular correlations may be written as:

$$W(\theta) = \sum_{k=0}^{K_{max}} A_K P_K(\cos \theta), \quad (4.6)$$

where $P_K$ is the $K$th Legendre polynomial and $K_{max}$ is two times the lesser of the intermediate spin or the angular momentum carried away by a $\gamma$ ray [88]. Figure 4.19 shows the angular correlations for several decay schemes studied in this work.

For $\beta\beta$ decay to excited states, only the $0^+$ and $2^+$ states may be populated. As such, only the $0^+ \rightarrow 2^+ \rightarrow 0^+$ and $2^+ \rightarrow 2^+ \rightarrow 0^+$ decay sequences are of interest.
The effects of the different decay modes may be accurately studied using GEANT4, as all the decay parameters are set by theory and the detector geometry is well known. The change in the efficiency of the apparatus is highly position dependent. This effect was simulated and factored into the efficiency curve shown in Fig. 4.20. The results were in good agreement with the measured EC decay of $^{102}$Rh to the $2^+_2$ state after corrections for the energy dependence are made.

4.4.6 Final efficiency

The coincidence efficiency is shown in Fig. 4.20 as a function of position. The efficiency was measured as a function of the radius in four directions (up/down/left/right). The plot shows the average of all four scans. The size of the $^{96}$Zr sample is shown for comparison.

The coincidence efficiency is fit to a function of the form

$$
\epsilon(r) = \frac{a}{1 + br^2 + cr^4}.
$$

(4.7)

The total efficiency involves the efficiency in the vicinity of both disks. In order to calculate this, we will actually calculate the product of the efficiency and the size of
Figure 4.20: (Color online) Coincidence efficiency as a function of \( r \) for the two-coaxial apparatus. The radii of the two samples is shown in blue and the holder in gray.

For the \(^{96}\text{Zr}\) sample:

\[
N_0 \epsilon = N_1 \frac{\int_0^{r_1} \epsilon(r) r \, dr}{\int_0^{r_1} r \, dr} + N_2 \frac{\int_{r_2}^{r_3} \epsilon(r) r \, dr}{\int_{r_2}^{r_3} r \, dr}.
\]  

(4.8)

Here, \( N_1 \) and \( N_2 \) are the number of \(^{96}\text{Zr}\) nuclei in the inner and outer sample, respectively. \( r_1 \) is the radius of the inner disk, while \( r_2 \) and \( r_3 \) are the radii of the outer annular disk.

The final error budget for this measurement is shown in Table 4.4. The largest source of error (3.1\%) comes from the calibrated \(^{137}\text{Cs}\) source that was used to determine the total detector efficiency, and thus limited the accuracy to which the activity of the \(^{102}\text{Rh}\) source could be measured. The second largest effect (2.5\%) comes from the uncertainty in the detector and source placement. As is seen by investigating the detector separation, inaccuracies as small as 1 mm in the separation distance can have a sizable effect on the efficiency. The only other sizable contribution to the error budget (2.4\%) stems from small asymmetries discovered when measuring the radial
Table 4.4: The systematic error budget for $\beta\beta$ decay of $^{96}$Zr. These uncertainties are for the decay to the first excited $0^+_1$ state. Note that uncertainty on the energy dependence and attenuation correction factors is energy dependent and will be higher for the decays to higher excited states.

<table>
<thead>
<tr>
<th>Uncertainty contribution</th>
<th>Percent error</th>
</tr>
</thead>
<tbody>
<tr>
<td>Calibration of $^{137}$Cs source</td>
<td>3.1 %</td>
</tr>
<tr>
<td>Energy dependence correction factor</td>
<td>0.4 %</td>
</tr>
<tr>
<td>Attenuation correction factor</td>
<td>0.2 %</td>
</tr>
<tr>
<td>Z dependence correction factor</td>
<td>0.15 %</td>
</tr>
<tr>
<td>Detector and source geometry</td>
<td>3.0 %</td>
</tr>
<tr>
<td>Non-symmetrical efficiency curve</td>
<td>2.4 %</td>
</tr>
<tr>
<td>Dead time</td>
<td>0.15 %</td>
</tr>
<tr>
<td>Uncertainty in $^{102}$Rh half-life</td>
<td>0.82 %</td>
</tr>
<tr>
<td><strong>Total uncertainty</strong></td>
<td><strong>5.0 %</strong></td>
</tr>
</tbody>
</table>

efficiency. As was previously noted, the results in Fig. 4.20 are the average of four scans along each axis. The detectors show some asymmetry in their efficiency, which is incorporated here through evaluations of the uncertainty in the fit of Fig. 4.20.

The final systematic error of 5.0% is smaller than that of [69], where it was reported at 6.8%. This is due to the GEANT4 simulation work, which is a new improvement on this pre-existing experiment.

4.5 $\beta$-decay results

The decay scheme for the $\beta$ decay of $^{96}$Zr is shown in Fig. 2.1. The excited-state transitions in the daughter nucleus, $^{96}$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 $\beta$ decay, since $T_{1/2}(^{96}$Zr) $\gg T_{1/2}(^{96}$Nb). The three most probable decay modes are given in Table 4.5. 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
Table 4.5: 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 $\gamma$ rays is given for each sequence.

<table>
<thead>
<tr>
<th>Decay sequence</th>
<th>$f_b$</th>
<th>$\gamma_1$</th>
<th>$\gamma_2$</th>
<th>$\gamma_3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$5_0^+ \rightarrow 4_1^+ \rightarrow 2_0^+ \rightarrow 0_0^+$</td>
<td>51.5%</td>
<td>568.9 keV</td>
<td>1091.3 keV</td>
<td>778.2 keV</td>
</tr>
<tr>
<td>$5_0^+ \rightarrow 3_0^+ \rightarrow 2_0^+ \rightarrow 0_0^+$</td>
<td>18.8%</td>
<td>460.0 keV</td>
<td>1200.0 keV</td>
<td>778.2 keV</td>
</tr>
<tr>
<td>$5_0^+ \rightarrow 4_0^+ \rightarrow 2_0^+ \rightarrow 0_0^+$</td>
<td>11.3%</td>
<td>849.9 keV</td>
<td>810.8 keV</td>
<td>778.2 keV</td>
</tr>
</tbody>
</table>

three coincident $\gamma$ rays, which differentiates the analysis from that of $\beta\beta$ decay in Sec. 4.3.

The region of interest for $\beta$ decay was chosen very carefully in order to maximize the signal-to-noise ratio while minimizing systematic uncertainties involved in calculating the detector efficiency for three coincidence $\gamma$ rays. The result is a search for events where two of the three $\gamma$ rays are detected in coincidence, with the third escaping the HPGe detectors. This is referred to as a two $\gamma$-ray event. This ROI is still a $\gamma - \gamma$ coincidence, which makes the efficiency directly comparable to the $^{102}$Rh measurements. As such, this ROI allows the efficiency to be calculated via GEANT4 simulations as discussed in the following section.

No cuts are made on the NaI annulus. The reason for this is that no direct measurements of the NaI efficiency have been performed. The efficiency of the NaI cut would necessitate simulations in GEANT4, where undesirable systematic uncertainties in the efficiency would arise. Furthermore, the trigger efficiency of the NaI is not included in the simulation and would be one of the largest contributors to the efficiency. Interestingly, GEANT4 simulations showed that 86-90% of these two $\gamma$-ray events resulted in a signal in the NaI annulus. The NaI cut would decrease the background roughly by a factor of two, for a total signal-to-noise improvement of $\approx 1.7$. As will be seen later, the background was found to be sufficiently low without this cut due to the coincidence between the two HPGe detectors and the excellent HPGe energy resolution.
Another option is to search for events where two $\gamma$ rays have complete energy deposition in one HPGe and the third $\gamma$ ray is detected in coincidence. This is referred to as a three $\gamma$-ray event. The summing of the two $\gamma$ rays in one detector was not experimentally measured and necessitates simulations adding additional systematic uncertainties. Furthermore, GEANT4 simulations show that this ROI has a factor of 5-6 smaller efficiency than the chosen ROI, as expected. The summing of the two peaks will result in a much lower background due to the higher energy event, although this is not enough to compensate for the reduced efficiency. In this case the NaI is used in anticoincidence, as full energy deposition occurs in the HPGe detectors, leaving no signal in the NaI annulus.

Histograms resulting from this analysis are shown in Fig. 4.21 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 $\gamma$-ray events, while the bottom three histograms show the search for three $\gamma$-ray events. The $\pm 2\sigma$ ROI and background estimation regions are also shown.

The results of this analysis are detailed in Table 4.6. The analysis is performed for the three decay modes detailed in Table 4.5. Each decay mode produces three $\gamma$ rays, resulting in three different two $\gamma$-ray events. There are three additional possible three $\gamma$-ray events. This gives 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 $\gamma$-ray coincidence resulting from neutron scattering on lead. The 460.0-778.2 keV coincidence was contaminated by the 463.0-911.2 keV $\gamma$-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.
Figure 4.21: The ROI histograms for a subset of the $^{96}$Zr $\beta$-decay coincidences. Histograms are shown for the $5_0^+ \rightarrow 4_1^+ \rightarrow 2_0^+ \rightarrow 0_0^+$ decay. The signal and background estimation regions are highlighted by the red and blue shading, respectively. 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.
Table 4.6: The results of the search for $^{96}$Zr’s $\beta$ decay are summarized. For each decay sequence and coincident pair we give the effective efficiency $\epsilon_{\text{eff}}$ and its associated systematic error $\sigma_{\text{syst}}$, number of observed counts in the ROI $N_{\text{obs}}$, and the expected background $N_{\text{bkgd}}$. The $N = 2$ cut was used. The efficiency and systematic error will be discussed further in Sec. 4.6.

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

* Sum of two coincident $\gamma$ rays

4.6 $\beta$-decay efficiency calculation

Given that these decays contain three coincidence $\gamma$ rays, the procedure for calculating the efficiency needs to be altered. The efficiency measurements were performed for two coincident $\gamma$ rays. When extending these efficiency measurements to three coincident $\gamma$ rays, the corrections become difficult to disentangle. This includes the angular correlation between the $\gamma$ rays, detection of two $\gamma$ rays in the same detector (known as accidental coincidence summing), and the efficiency of the NaI annulus. These issues are resolved by using GEANT4 simulations of the efficiency normalized to the $^{102}$Rh efficiency

$$
\epsilon(96\text{Zr}) = \epsilon_{\text{sim}}(96\text{Zr}) \frac{\epsilon_{\text{exp}}(102\text{Rh})}{\epsilon_{\text{sim}}(102\text{Rh})}.
$$

(4.9)
Although this may seem heavily reliant on GEANT4 at first, most of the corrections in the previous section were performed using GEANT4 to extreme accuracy. In fact, the only correction previously done using experimental data and now done using GEANT4 is the energy dependence of the efficiency, which is shown in Fig. 4.15 to have excellent agreement. The correction factor Eq. (4.2) was calculated using both the measured and simulated efficiency curves for all possible γ-ray pairs resulting from $^{96}$Zr’s β decay. The correction factor differed by $< 1.4\%$ between the simulated and measured values, with the higher discrepancy occurring for the higher energy γ rays. This difference was added as systematic uncertainty for the β-decay efficiency calculation.

In order to carry out the simulation, the NaI annulus was added into the GEANT4 geometry, as shown in Fig. 4.22. This was necessary to model backgrounds caused by signal γ rays Compton-back scattering from the NaI into the HPGe detectors. It also allows for investigation of signals within the NaI, such as a triple coincidence between the two detectors and the NaI. The NaI annulus was not needed in the earlier simulations because the signal region required complete energy deposition in the HPGe detectors. As such it was removed in order to reduce computation time.
The regions of interest for $\beta$ decay are still coincidences where both detectors are triggered. This is important because the trigger efficiency is identical for the investigated ROI and the efficiency measurements. The trigger of the HPGe detectors is not included in the GEANT4 simulation and would introduce many uncertainties when calculating the efficiency via simulation. Instead, we are left with the interaction probability of the third $\gamma$ ray as the largest uncertainty. The angular distribution, interaction cross section of $\gamma$ rays at these energies with these materials, and detector geometry are all well known, so the simulation is well equipped to deal with the third $\gamma$ ray. In two $\gamma$-ray events, the probability of the third $\gamma$ ray escaping the HPGe detectors leads to a small correction factor on the order of $1 - \epsilon_p$. Here, $\epsilon_p$ is the efficiency for a $\gamma$ ray depositing full or partial energy in the detector. When searching for complete energy deposition of the three $\gamma$ rays, the correction factor is on the order of $\epsilon$, the HPGe detection efficiency for a single $\gamma$ ray. The uncertainty on the single HPGe detector efficiency $\sigma_\epsilon$ in the GEANT4 simulation 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.

Some results of this simulation are shown by the efficiency curves in Fig. 4.23. Each of the ROIs in Table 4.6 has its own efficiency curve, only three of which are shown here. The effective efficiency $\epsilon_{eff}$ in Table 4.6 is the integrated product of $\epsilon(r)$ and the sample enrichment, given by Eq. (4.8), divided by the total number of $^{96}$Zr nuclei $N_0 = N_1 + N_2$. Interestingly, the efficiency curve does not decrease as sharply with respect to $r$. The efficiency for complete detection of all three $\gamma$ rays is much lower than that of the two $\gamma$-ray events. This is expected as the detection efficiency $\epsilon_f$ of two $\gamma$ rays is $\epsilon_f \propto \left(\frac{3}{2}\right) \epsilon^2 (1 - \epsilon_p)$, while detection of three $\gamma$ rays is $\epsilon_f \propto \epsilon^3$.

A summary of systematic errors is shown in Table 4.7. These systematic uncertainties are added in quadrature in addition to the previously discussed systematic uncertainties.
Figure 4.23: (Color online) The efficiency as a function of position for $^{96}$Zr’s $\beta$ decay. The decay contains three coincident $\gamma$ rays (568.9, 1091.3, and 778.2 keV). The efficiency is shown for events given observation of a 1091.3 keV $\gamma$ ray in one detector. The other detector will see peaks at 568.9, 778.2, and $568.9 + 778.2 = 1347.1$ keV.

uncertainties from the $\beta\beta$-decay calculations in Table 4.4. The efficiency of a single HPGe detector and energy dependency of the efficiency contributions have already been discussed. These two contributions differ as one is a measure of the overall normalization of the Monte Carlo and the second is a result of the Monte Carlo. The final uncertainty, the statistical uncertainty of the GEANT4 simulations, results from fitting the efficiency curve in Fig. 4.23 to Eq. (4.7). This could be improved by increasing the number of simulated events. Doing so would require increased computational time, but as the Monte Carlo is normalized to experimental data and uses a functional fit, the contribution to the systematic uncertainty would not necessarily converge to zero uncertainty.
Table 4.7: The systematic error budget for the $\beta$ decay of $^{96}$Zr. The $\beta\beta$-decay efficiency comes from Table 4.4. 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.

<table>
<thead>
<tr>
<th>Uncertainty contribution</th>
<th>Percent error</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\beta\beta$ decay efficiency</td>
<td>5.0 %</td>
</tr>
<tr>
<td>Total efficiency of HPGe in GEANT4</td>
<td>5.5 %</td>
</tr>
<tr>
<td>Energy dependence of efficiency in GEANT4</td>
<td>0.92 %</td>
</tr>
<tr>
<td>Angular distributions</td>
<td>0.30 %</td>
</tr>
<tr>
<td>Statistical uncertainty of GEANT4</td>
<td>0.61 %</td>
</tr>
<tr>
<td>Total uncertainty</td>
<td>7.5 %</td>
</tr>
</tbody>
</table>
This chapter details the analysis of all the data collected by the two-clover apparatus. The $^{156}$Dy source was in place during this time, allowing limits to be set on the $E^*E^*E^*C$ decay of $^{156}$Dy to a number of excited final states. The analysis in this chapter will follow roughly the same outline as Chapter 4, as both apparatuses exhibit similar features and drawbacks.

5.1 Data collection

Data collection was performed the same way as documented for the two-coaxial HPGe apparatus. New runs were started every one to five days, with an average length of three days. Two data sets were collected: one with only $^{156}$Dy$_2$O$_3$ sample #1 present (150.95 mg $^{156}$Dy) and one with both enriched samples present (213.57 mg $^{156}$Dy). These sets are referred to as data sets #1 and #2 respectively. Data set #1 contains 99.13 days of data collection and data set #2 contains 132.82 days of data collection.

Each individual run was closely checked for gain changes and drifts in each de-
tector segment. This was done by plotting the spectra as a function of time. Any runs in which peaks were seen to migrate by more than one channel were removed and are not included in the present analysis. The quality control tests performed on the clover detectors were identical to those performed on the two coaxial apparatus.

5.2 Data processing

The ADC was calibrated to energy for each run using four naturally occurring background $\gamma$ rays: 238.6, 511.0, 911.2, and 1460.8 keV. This calibration was done for each individual clover segment. An additional quantity, referred to as the ‘addback’ energy, was also calculated for each clover detector. The addback energy was calculated for events where two or more segments fired in one clover detector and is the sum of the energy deposited in those segments. A 30 keV threshold was used to determine whether or not a segment fired. This threshold was chosen to be comfortably above the 10 keV noise pedestal of the detectors and electronics while still allowing low energy Compton scatter events. Analysis showed that lowering the threshold further did not have a significant effect on the addback efficiency. In cases where only one detector segment in a clover fires, this event is recorded as the ‘singles’ energy.

The addback energy was again calibrated to the same four peaks as used in the initial calibration. This results in a very small correction to the energy calibration. This secondary calibration is necessary because the addback energy resolution is equal to the energy resolution of the two contributing segments added in quadrature, or a factor of $\approx \sqrt{2}$ worse, as shown in Fig. 5.1. The second calibration corrects for any shifts that may have been caused by the decreased resolution. The results of the data processing are shown in Fig. 5.2. Spectra are shown for a clover detector in singles and addback mode, with and without the NaI annulus veto.

After the clover detectors were moved underground, it was noticed that segment
Figure 5.1: (Color online) The energy dependent resolution of both clover detectors over the entirety of runs with $^{156}\text{Dy}$ sample #1 in place. The resolution is shown for the detectors operated in both addback and singles mode. The functional fits are shown by the black lines. Note $FWHM \approx 2.355 \sigma$.

Figure 5.2: (Color online) The energy spectrum as seen by the clover detectors. The spectra is shown for a clover detector in both singles and addback mode, with and without the NaI annulus veto. Notice that the addback spectra falls off dramatically below 200 keV. This is expected as low energy events will rarely Compton scatter between clover segments. Similarly, the singles and addback spectra begin to converge at higher energies where Compton scattering is more prominent.

three of clover one, hereinafter referred to as c13, had an irreducible electronic noise present, decreasing the segment’s resolution by a factor of $\approx 3$. This presents multiple problems. Firstly, broader peaks make it harder to accurately calibrate the energy spectra and perform data quality checks. Furthermore, background peaks can bleed into the ROI with the decreased resolution. This effect can be combated by using stricter energy cuts, say $\pm 1\sigma$, but will result in a sizable decrease in efficiency. As
such, events in which segment c13 triggered were removed from the present analysis. This is equivalent to using this one segment in anticoincidence. The efficiency reduction of this cut is shown in Sec. 5.4.

5.3 Data analysis

The present analysis searches for coincident $\gamma$ rays. When using the clover detectors, $\gamma - \gamma$ coincidences may be one of two types: internal or external. External coincidences occur when both clover detectors detect a $\gamma$-ray interaction. Internal coincidences, on the other hand, occur between two segments of the same clover detector when the other clover detector does not record any events. By searching for both internal and external coincidences, the efficiency of the $\gamma - \gamma$ coincidence technique is increased. This can be seen by inspecting the angular distribution of coincident $\gamma$ rays $W(\theta)$, shown in Fig 4.19. The two-coaxial HPGe apparatus, and external coincidences in the two-clover apparatus, mainly cover the $\theta \sim \pi$ rad region. By including internal coincidences, the clover detectors are sensitive to the $\theta \sim 0$ rad region as well. As Compton scattering is much more likely between two adjacent segments than between the two separate clover detectors, the internal coincidence spectra will have a higher background.

Each of the decay states listed in Table 2.2 was investigated separately. In the following sections, the specific analysis criteria are discussed along with the accompanying resultant histograms. In all cases, the energy cuts used were $\pm 2\sigma$. Furthermore, the same algorithms that were used for determining the minimum detectable signal above background for the $^{96}$Zr decays were employed here. The background estimation region was always taken as the largest peak-free region surrounding the ROI, ideally a $\pm100$ keV range around the ROI.
5.3.1 To the 1946.4 keV state

As can be seen in Fig. 5.3, the 1946.4 keV state will primarily decay via two modes. The most intense transition creates a 1857.4 + 88.97 keV coincidence, which will be studied in this analysis. Somewhat unfortunately, the only other intense transition produces a single $\gamma$ ray and is therefore not suited for the coincidence technique used in the present experiment.

Given that the 88.97 keV $\gamma$ ray is very low in energy, it will rarely Compton scatter between detector segments. The first requirement of the analysis is a $88.97 \pm 2\sigma$ keV event in one detector segment. External coincidences then require that this 88.97 keV $\gamma$ ray is the only event in its clover detector and a 1857.4 keV $\gamma$ ray is detected in the opposing detector. Given the higher energy of the 1857.4 keV $\gamma$ ray, both addback and singles events are included in the search. Internal coincidences require the 1857.4 keV $\gamma$ ray to be observed in a segment of the same clover and no triggers in the remaining six clover segments. Addback is not attempted for internal coincidences as the efficiency becomes difficult to calculate without heavily relying on Monte Carlo techniques. A sensitivity study was performed to optimize the ROI. Although the internal coincidence background is higher and its efficiency lower, the most stringent limits are found by including both internal and external coincidences.
The ROI spectra found from the sum of internal and external coincidences is shown in Fig. 5.4.

The resulting spectra bears much similarity to those of the $^{96}$Zr analysis. Although the two-clover apparatus has more radioactive impurities, and therefore a higher background level, an extremely low background ROI is achieved through the coincidence technique. The high $Q$ value of $ECEC$ and high energy $\gamma$-ray transitions help to further reduce the background.

5.3.2 To the 1952.4 keV state

The decay sequence for the 1592.4 keV state is shown in Fig. 5.5. A 709.9+1242.5 keV $\gamma$-ray coincidence is produced in 44.7% of decays. The only other intense decay mode results in three coincidence $\gamma$ rays with a branching ratio of 46.0%.

As the efficiency measurements were performed with two $\gamma$-ray coincidences, the two $\gamma$-ray decay will dictate the analysis cuts. For this decay, the energy of both coincident $\gamma$ rays is high enough that a significant portion will Compton scatter between clover segments. As such, the analysis procedure differs slightly from the one
Figure 5.5: (Color online) Decay scheme for the $0^{-} 1952.4$ keV state of $^{156}$Gd following $ECEC$ to a resonant state. Only the most intense transitions are shown, while all other transitions have a branching ratio $< 3.8\%$.

Figure 5.6: (Color online) The ROI spectra for $ECEC$ to the 1952.4 keV state including both runs #1 and #2. Events in coincidence with 709.9 keV are shown. The background estimation regions are indicated by the blue shading and the signal region, 1242.5 keV, by the red shading. The red curve shows the background level and the minimum detectable signal at the 90% confidence level.

outlined in the previous section. External coincidences search for the 709.9+1242.5 keV coincidence between the two clover detectors operating in both singles and add-back mode. This differs from the previous section where the 88.97 keV $\gamma$ ray was only detected in singles mode. Internal coincidences, once again, are between two detector segments of the same clover with no addback corrections made. This same analysis procedure will be used for the other ROIs with two coincident $\gamma$ rays above 400 keV.
The results of this analysis procedure are shown in Fig. 5.6. The most statistically sensitive ROI was found by triggering off of the 709.9 keV event and investigating the corresponding region around 1242.5 keV.

This analysis procedure does not intrinsically distinguish the two and three $\gamma$-ray decay modes from the 1952.4 keV state. For example, it is possible that the 1153.5 and 88.97 keV $\gamma$ rays are detected in one clover then re-interpreted as a 1242.5 keV addback event. This possibility increases the experimental sensitivity of this ROI by including an additional decay mode. The increase will be discussed further in the efficiency calculations.

5.3.3 To the 1988.5 keV state

This nuclear state is of particular interest because it is the only $0^+$ state available for a resonant $ECEC$ transition. Somewhat unfortunately, the state suffers from a dearth of nuclear data. No data is present on the possible decay modes from this state. Although the nature of the excitation will dictate the decay modes, it is reasonable to assume that the $0^+_4 \rightarrow 2^+_0 \rightarrow 0^+_{g.s.}$ transition will be one of the strongest, if not the most intense transition. This is true for the other excited $0^+$ states in $^{156}$Gd. As such, we search for events matching this decay mode and producing an 1899.5 + 88.9 keV $\gamma$-ray coincidence.

The analysis was performed identically to that of the 1946.4 keV state outlined in Sec. 5.3.1, with the 1857.4 keV ROI replaced with 1899.5 keV. The results are shown in Fig. 5.7.

5.3.4 To the 2003.7 keV state

This $2^+$ state has a more complicated decay scheme than the other studied states. The state has been observed to decay to six different states, with the most intense decay sequence only accounting for 23% of all decays. As our efficiency measurements
Figure 5.7: (Color online) The ROI spectra for ECEC to the 1988.5 keV state including both runs #1 and #2. Events in coincidence with 88.97 keV are shown. The background estimation regions are indicated by the blue shading and the signal region, 1899.5 keV, by the red shading. The red curve shows the background level and the minimum detectable signal at the 90% confidence level.

were performed for two coincidence $\gamma$ rays, we limit ourselves to those cases, all of which are shown in Fig. 5.8. Unfortunately, the three decays producing only two $\gamma$ rays only have a combined branching ratio of 25.8%. Two of these three decays has an additional decay mode via the 88.97 keV $2^+$ state, producing three coincidence $\gamma$ rays. By including these ternary decays, the total branching ratio is nearly doubled to 46.2%.

The procedure outlined in Sec. 5.3.2 is again followed for these three decay modes. The 684.0+1319.7, 7361.3+1242.5, and 849.6+1154.2 keV coincidences are investigated, with the results shown in Fig. 5.9. After the addition of sample #2, the background rate for the decay modes progressing via the 1242.5 and 1154.2 keV states was found to have an increased background. It was easily realized that the internal coincidence channel was responsible for the majority of the background increase. Although the exact source of the background is unknown, it is not unreasonable to assume contaminants in the sample #2 are responsible. For the case of the decay to the $2^+$ 1154.2 keV state, a better limit was produced by only investigating external coincidences. In this case, the loss in efficiency is compensated by the decreased
Figure 5.8: (Color online) Decay scheme for the $2^+$ 2003.7 keV state of $^{156}$Gd following ECEC to a resonant state. Only the most intense two $\gamma$-ray transitions are shown, while all other two $\gamma$-ray transitions have branching ratio $< 0.5\%$. The three $\gamma$-ray transitions associated with these two $\gamma$-ray transitions are also shown.

background.

5.4 Efficiency measurements

Efficiency data were taken using a variety of point sources: $^{133}$Ba, $^{56}$Co, $^{60}$Co, $^{137}$Cs, $^{152}$Eu, $^{22}$Na, and $^{54}$Mn. Once again, in order to measure the coincidence efficiency, a $^{102}$Rh source was used. This source was produced using the $^{102}$Ru($p$, $n$)$^{102}$Rh reaction with 5 MeV protons at the TUNL tandem accelerator. This was a different source than the one used for the efficiency measurements on the coaxial HPGe detectors. The source was cut into a piece 1.65 mm in diameter and 1.42 mm in height with an activity of $1.26 \pm 0.04$ kBq. This low activity was chosen such that the count rate would be low, and thus the dead-time corrections would be negligible. High statistics were then obtained by counting for $> 1$ day at different positions along the detector faces. The efficiency data were analyzed using the same analysis code as was used for the $^{156}$Dy analysis. Only the ROI and detector resolution were changed to accurately reflect the run conditions.

In discussing the efficiency of the clover detectors, a simple coordinate system is adopted. The origin is centered in between the two clover detectors with the $+z$-axis
Figure 5.9: (Color online) The ROI spectra for $E_{CEC}$ to the 2003.7 keV state including both runs #1 and #2. Events are shown in coincidence with 1319.7, 1242.5, and 849.6 keV respectively. The background estimation regions are indicated by the blue shading and the signal region by the red shading. The red curve shows the background level and the minimum detectable signal at the 90% confidence level.
pointing towards the center of clover detector one's face. The $x$-axis runs parallel to the floor and along the edge between two segments. Similarly, the $+y$-axis points towards the ceiling and also runs along the edge in between two segments. Notably, $c_{13}$ occurs in the $(+x, +y, +z)$ quadrant.

One might expect the efficiency of the two-clover apparatus to be highly position dependent, possibly peaking at the center of the individual crystals with a lull at the origin, where there is no crystal present. In fact, the coincident detection efficiency was found to peak at the origin, and is relatively constant from $-1.5 < x < 1.5$ cm, as is shown in Fig. 5.10. This motivated the size and placement of the Dy$_2$O$_3$ samples. The reason that these detectors have a constant efficiency at the origin is shown in Fig. 5.11, where the efficiency of the four $x < 0$ segments balances the efficiency of the four $x > 0$ segments. The summed efficiency, shown in Fig. 5.10 is therefore very symmetric about the origin. The efficiency in Fig. 5.10 appears to fall off faster when measured along the $y = x$ line. This is because the graph is plotted as a function of position on the $x$-axis, when the true distance from the origin is a factor of $\sqrt{2}$ larger.

The internal efficiency of clover two will not change with the omission of $c_{13}$. On the other hand, the internal efficiency of clover one and the external efficiency become very asymmetric after the removal of $c_{13}$, as shown in Fig. 5.12. From these plots, it is evident that the internal efficiency falls off much faster away from the origin than the external efficiency. The omission of $c_{13}$ significantly lowers the efficiency in quadrant one, although much more so for internal coincidences than external coincidences. This is expected as for external coincidences we are only omitting one of the two detectors in this quadrant, while for internal coincidences in clover one we are omitting all of the detectors in this quadrant.

The final efficiency was calculated by integrating the two-dimensional efficiency
Figure 5.10: (Color online) The average coincident efficiency of the two-clover apparatus is shown along the two symmetry axes: $y = 0$ and $y = x$. The efficiency is shown for external coincidences, internal coincidences, and addback coincidences. These efficiencies are for the $^{102}$Rh ROI and include $c_{13}$. The maximum dimension of the $^{156}$Dy sample is shown by the cream box, with the other dimensions of the rectangular sample being smaller.

Figure 5.11: (Color online) The coincident efficiency of the two-clover apparatus as measured along the $x$-axis. The combined efficiency of the four segments positioned at $x < 0$ is shown in the left panel and the combined efficiency of the four $x > 0$ segments is shown in the right panel. The black points represent the external-coincidence efficiency and the red points represent the internal-coincidence efficiency. Note that this analysis includes $c_{13}$.
Figure 5.12: (Color online) The coincident efficiency of the two-clover apparatus as a function of position with the omission of $c13$ is shown. The three panels show, from left to right, the internal efficiency of clover one, the internal efficiency of clover two, and the external efficiency. Note that the color shading is relative for each plot and does not represent the overall efficiency difference between the three detection modes.

Table 5.1: Coincident efficiency $\epsilon_{\gamma\gamma}$ of the two-clover apparatus as measured with $^{102}$Rh. All values are given in [%].

<table>
<thead>
<tr>
<th>Sample</th>
<th>Clover 1 Internal</th>
<th>Clover 2 Internal</th>
<th>External</th>
</tr>
</thead>
<tbody>
<tr>
<td>#1</td>
<td>0.123 ± 0.002</td>
<td>0.242 ± 0.008</td>
<td>0.548 ± 0.003</td>
</tr>
<tr>
<td>#2</td>
<td>0.127 ± 0.002</td>
<td>0.248 ± 0.008</td>
<td>0.557 ± 0.004</td>
</tr>
</tbody>
</table>

over the sample size

$$\epsilon_{\gamma\gamma} = \frac{1}{wh} \int_{-w/2}^{w/2} \int_{-h/2}^{h/2} \epsilon_{\gamma\gamma}(x, y) dy dx,$$

where $w$ and $h$ are the width and height of the sample, respectively. The integration was performed for all three efficiency plots and both $^{156}$Dy samples and is summarized in Table 5.1. Of course, a number of corrections are necessary in order to apply these $^{102}$Rh efficiency measurements to the possible $^{156}$Dy decay modes. These corrections will now be discussed in detail.

5.4.1 Energy dependence

The energy dependence of the efficiency was again measured. As one of the $\gamma$ rays of interest for the ECEC decay of $^{156}$Dy has a relatively low energy, 88.97 keV, special care must be taken in calculating the efficiency of the detector at low energies. The
Figure 5.13: (Color online) A GEANT4 model of a clover HPGe detector. The crystals are shown in green, with the aluminum shell in blue and end cap window in red. A head-on view of the crystal geometry without the shell is shown to the right.

Figure 5.14: (Color online) Singles efficiency as a function of energy for both clover detectors. Simulation data were produced using GEANT4. The functional fit is in black. Note that the efficiency of clover one is $\approx 25\%$ lower due to the omission of segment three.

clover detector has a 2.54 mm aluminum front window which will attenuate $\gamma$ rays below $\approx 100$ keV. In order to carefully account for this effect, GEANT4 simulations of the clover efficiency were performed at low energies. The GEANT4 model of the clover detector geometry is shown in Fig. 5.13. The results of this simulation and the experimental efficiency measurements are shown in Fig. 5.14. The efficiency peaks around 80 keV, before sharply decreasing due to attenuation by the aluminum window. The efficiency found using this procedure is roughly the same as the efficiency measured using the 121.8 keV peak from $^{152}$Eu, thus substantiating the Monte Carlo with experimental data.
The efficiency was fit to a function of the form

$$\epsilon_\gamma(E) = a + b \exp(c \log E + d \log^2 E). \quad (5.2)$$

This equation is equivalent to Eq. (4.1) with an additional $\log^2(E)$ term to model the efficiency drop off at low energies. The correction was calculated as a ratio of the coincidence energy for $^{102}$Rh and the $^{156}$Dy ECEC ROI, as given by Eq. (4.2). In calculating these corrections, the data points resulting from the GEANT4 simulations were only used for evaluation of the 88.97 keV efficiency. All other efficiencies were calculated using only the experimental data and the reduced form of $\epsilon_\gamma(E)$ given by Eq. (4.1). The final correction factors, for each of the studied transitions, is shown in Table 5.2.

5.4.2 Addback factor

Following the analysis of Ref. [74], the total efficiency $\epsilon_T$ may be written as

$$\epsilon_T = \epsilon_S + \epsilon_{AB} = \epsilon_S \times (1 + f), \quad (5.3)$$

where $\epsilon_S$ is the detector efficiency in singles mode and $\epsilon_{AB}$ is the detector efficiency in addback mode. The addback fraction $f$ was measured using several point sources and is shown in Fig. 5.15. The source’s activity will cancel out in measurements of the addback fraction, allowing $f$ to be measured using several different sources without increased systematic uncertainties. The addback fraction is then fit to a function of the form

$$f(E) = a + b \log(E) + c \log^2(E) \quad (5.4)$$

in order to interpolate $f(E)$.

The addback factor was measured at a distance of 25 cm in order to prevent accidental summing of coincident $\gamma$ rays. The experiment on $^{156}$Dy, however, was conducted at a distance of 1.5 mm. As such, there are two addback corrections: the
Figure 5.15: (Color online) The addback ratio for the two clover detectors. Data are shown by the points and the functional fit is shown by the solid curves. The addback factor in clover one is noticeably lower due to the omission of segment three.

The addback factor $f(E)$ and a correction arising from the position dependence $F(z)$. Although both of these factors are energy dependent, $f(E)$ has a much stronger energy dependence than $F(z)$ at the considered energies. As such, $f(E)$ was measured at 25 cm using a variety of sources, and $F(z)$ was measured at 1.5 mm using fewer sources: those producing single $\gamma$ rays and the $^{102}$Rh source. These measurements were confirmed by GEANT4 simulations, which are shown in Fig. 5.16. $F(z)$ ranged from 1.25 to 1.35 over the energy range studied.

The total addback factors are calculated as the product of $F(z)f(E)$ and increase the total clover detection efficiency by a factor of $F(z)f(E) + 1$. The combined addback factors are given in Table 5.2. It should be noted that this correction factor only increases the external coincidence detection efficiency, as no addback detection was utilized for internal coincidences. For the two states producing the 88.97 keV $\gamma$ ray (1946 and 1988 keV), the addback factor is considerably lower. This is because the 88.97 keV $\gamma$ ray is a detected as singles event and only one detector is operated
Figure 5.16: (Color online) The improvement factor $F(z)$ in the addback ratio as a function of energy. The improvement factor is given as the ratio $f(E, z = 1.5 \text{ mm})/f(E, z = 25 \text{ cm})$. The functional fit is also shown.

in addback mode. For the other transitions, both detectors are operated in addback mode, essentially doubling the efficiency increase due to addback.

5.4.3 Target attenuation

As the samples were very small and only stored in polyethylene bags, $\gamma$-ray attenuation due to the sample is expected to be very small. Once again, the target attenuation was calculated using GEANT4. The samples were modeled in GEANT4 and events were simulated with and without the sample present. As expected, the attenuation was relatively small, $\approx 1 - 2\%$, for all decay modes with high-energy $\gamma$ rays. The two decay modes producing the 88.97 keV $\gamma$ ray, however, show a sizable attenuation factor: $\approx 14.2\%$ for run #1 and $\approx 28.9\%$ for run #2. This correction factor was calculated separately for internal and external coincidences and for both runs. Numerical values of the attenuation factor are given in Table 5.2 for run #2.

5.4.4 Detector separation and source geometry

The separation distance between the detectors was kept as small as possible in order to maximize the solid angle covered by the detectors. The same detector spacing, 1.8 mm, was used for the $^{102}\text{Rh}$ source measurements and for run #1 (including
Table 5.2: Efficiency correction ratios for different coincidences in $^{156}$Dy. The attenuation ratio of the $^{156}$Dy sample, the energy dependence ratio of the detectors, and the combined addback factor $F(z)f(E) + 1$ are all given. The attenuation values are given for run #2. All corrections are calculated as the efficiency-weighted average over all internal and external coincidences excluding c13.

<table>
<thead>
<tr>
<th>$J^\pi$</th>
<th>$\gamma_1$ [keV]</th>
<th>$\gamma_2$ [keV]</th>
<th>Sample attenuation $\epsilon(\gamma_1, \gamma_2)/\epsilon(468, 475)$</th>
<th>$F(z)f(E) + 1$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1$^-$</td>
<td>1857.4</td>
<td>88.97</td>
<td>0.713 ± 0.001</td>
<td>0.876 ± 0.004</td>
</tr>
<tr>
<td>0$^-$</td>
<td>709.9</td>
<td>1242.5</td>
<td>0.968 ± 0.002</td>
<td>0.333 ± 0.002</td>
</tr>
<tr>
<td>0$^+$</td>
<td>1899.5</td>
<td>88.97</td>
<td>0.725 ± 0.001</td>
<td>0.860 ± 0.004</td>
</tr>
<tr>
<td>2$^+$</td>
<td>684.0</td>
<td>1319.7</td>
<td>0.990 ± 0.002</td>
<td>0.328 ± 0.002</td>
</tr>
<tr>
<td>2$^+$</td>
<td>761.3</td>
<td>1242.5</td>
<td>0.989 ± 0.002</td>
<td>0.316 ± 0.002</td>
</tr>
<tr>
<td>2$^+$</td>
<td>849.6</td>
<td>1154.2</td>
<td>0.984 ± 0.002</td>
<td>0.307 ± 0.002</td>
</tr>
</tbody>
</table>

only sample #1). GEANT4 was utilized to calculate any difference in efficiency that occurred from the detector spacing and the physical extent of the $^{156}$Dy sample. The $^{102}$Rh source was determined to have a slightly asymmetric activity, possibly from its alignment in the beam. This effect was noticed and measured for by rotating the $^{102}$Rh source. Further simulations were performed in order to determine any effect on the efficiency measurements. Combining this effect with the physical thickness of the $^{102}$Rh and $^{156}$Dy sources gives a small correction to the efficiency: the internal coincidence efficiency of clover one increases 1.78%, clover two decreases by 3.49%, and the external coincidence efficiency decreases by 0.71%. The insertion of sample #2 increased the distance between the detectors to 3.2 mm and resulted in a small decrease in efficiency, 3.32 ± 0.01%, as calculated through the Monte Carlo.

5.4.5 Angular dependence of coincident $\gamma$ rays

As the efficiency measurements were performed using a $0^+ \rightarrow 2^+ \rightarrow 0^+$ decay sequence, corrections must be made for the various decay modes of interest with different spin assignments. The angular distribution between two coincident $\gamma$ rays is shown in Fig. 4.19 for a number of decays. The effect was accurately modeled using GEANT4. Table 5.3 shows the results of this Monte Carlo study. Interestingly, the
Table 5.3: The efficiency of the two-clover apparatus is given, in [%], for a subset of the different decay sequences studied. This efficiency given is for run #2, when both samples were in place.

<table>
<thead>
<tr>
<th></th>
<th>$0^+ \rightarrow 2^+ \rightarrow 0^+$</th>
<th>$1^- \rightarrow 2^+ \rightarrow 0^+$</th>
<th>$0^- \rightarrow 1^+ \rightarrow 0^+$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Clover 1 Internal</td>
<td>0.124</td>
<td>0.150</td>
<td>0.136</td>
</tr>
<tr>
<td>Clover 2 Internal</td>
<td>0.244</td>
<td>0.293</td>
<td>0.270</td>
</tr>
<tr>
<td>External</td>
<td>0.551</td>
<td>0.507</td>
<td>0.536</td>
</tr>
<tr>
<td>Total</td>
<td>0.919</td>
<td>0.950</td>
<td>0.942</td>
</tr>
</tbody>
</table>

Table 5.4: The final efficiency, in [%] for the $^{156}$Dy ROIs for both runs. The external coincidence, internal coincidence, and the total (combined) efficiency are all given.

<table>
<thead>
<tr>
<th>$J^g$</th>
<th>$\gamma_1$ [keV]</th>
<th>$\gamma_2$ [keV]</th>
<th>Run #1 $\epsilon_{ext}$</th>
<th>Run #1 $\epsilon_{int}$</th>
<th>Run #1 $\epsilon_{tot}$</th>
<th>Run #2 $\epsilon_{ext}$</th>
<th>Run #2 $\epsilon_{int}$</th>
<th>Run #2 $\epsilon_{tot}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$1^-$</td>
<td>1857.4</td>
<td>88.97</td>
<td>0.597</td>
<td>0.335</td>
<td>0.933</td>
<td>0.478</td>
<td>0.277</td>
<td>0.755</td>
</tr>
<tr>
<td>$0^-$</td>
<td>709.9</td>
<td>1242.5</td>
<td>0.373</td>
<td>0.135</td>
<td>0.508</td>
<td>0.353</td>
<td>0.131</td>
<td>0.484</td>
</tr>
<tr>
<td>$0^+$</td>
<td>1899.5</td>
<td>88.97</td>
<td>0.639</td>
<td>0.275</td>
<td>0.915</td>
<td>0.523</td>
<td>0.228</td>
<td>0.751</td>
</tr>
<tr>
<td>$2^+$</td>
<td>684.0</td>
<td>1319.7</td>
<td>0.362</td>
<td>0.142</td>
<td>0.504</td>
<td>0.340</td>
<td>0.136</td>
<td>0.476</td>
</tr>
<tr>
<td>$2^+$</td>
<td>761.3</td>
<td>1242.5</td>
<td>0.348</td>
<td>0.141</td>
<td>0.489</td>
<td>0.326</td>
<td>0.136</td>
<td>0.461</td>
</tr>
<tr>
<td>$2^+$</td>
<td>849.6</td>
<td>1154.2</td>
<td>0.343</td>
<td>0.131</td>
<td>0.474</td>
<td>0.323</td>
<td>0.128</td>
<td>0.451</td>
</tr>
</tbody>
</table>

external efficiency is lower for the more isotropic decays; however, the internal efficiency increases to compensate for this effect. This is likely from the minima that occurs at $\pi/4$ rad in the angular distribution for $0^+ \rightarrow 2^+ \rightarrow 0^+$.

5.4.6 Final efficiency

The final efficiency for each of the decay modes is summarized in Table 5.4. These results include all of the previously mentioned corrections. Additionally, the final error budget is shown in Table 5.5. This table has the uncertainties associated with each of the previous corrections, and bears much resemblance to the systematic error budget for the $^{96}$Zr experiment. Noticeably, an additional uncertainty due to the addback factor is included.

5.4.7 Contributions from three-$\gamma$ decays

As has already been mentioned, the decays to the 1952.4 and 2003.7 keV states have significant branching ratios for transitions that produce three coincident $\gamma$ rays. It
Table 5.5: The systematic error budget for the $ECEC$ decay of $^{156}$Dy to the 1946 keV state. Note that uncertainty on the energy dependence, attenuation, and addback correction factors are energy dependent and vary for each ROI.

<table>
<thead>
<tr>
<th>Uncertainty contribution</th>
<th>Percent error</th>
</tr>
</thead>
<tbody>
<tr>
<td>Calibration of $^{137}$Cs source</td>
<td>3.1 %</td>
</tr>
<tr>
<td>$^{102}$Rh efficiency measurements</td>
<td>1.2 %</td>
</tr>
<tr>
<td>Detector and source geometry</td>
<td>3.0 %</td>
</tr>
<tr>
<td>Angular dependence of $\gamma$ rays</td>
<td>1.20 %</td>
</tr>
<tr>
<td>Addback correction factors</td>
<td>2.9 %</td>
</tr>
<tr>
<td>Energy dependence correction factor</td>
<td>0.5 %</td>
</tr>
<tr>
<td>Attenuation correction factor</td>
<td>0.20 %</td>
</tr>
<tr>
<td>Z dependence correction factor</td>
<td>0.20 %</td>
</tr>
<tr>
<td>Dead time</td>
<td>0.20 %</td>
</tr>
<tr>
<td>Uncertainty in $^{102}$Rh half-life</td>
<td>0.82 %</td>
</tr>
<tr>
<td><strong>Total uncertainty</strong></td>
<td><strong>5.6 %</strong></td>
</tr>
</tbody>
</table>

is possible for a clover detector to detect two of these $\gamma$ rays in coincidence and reconstruct them as an addback event with the same energy as one of the two original $\gamma$ rays of interest. This is a signal event that will pass the analysis procedure utilized. As the apparatus will detect both of these decay modes, this effect will further increase the experimental sensitivity. As has already been seen in the single-$\beta$ decay of $^{96}$Zr, the efficiency of a triple coincidence is considerably lower than that of a double coincidence. The three $\gamma$-ray efficiency was simulated using GEA NT4. Using these simulations, the ratio $\epsilon_{3\gamma}/\epsilon_{2\gamma}$ was calculated. The ratio $\epsilon_{3\gamma}/\epsilon_{2\gamma}$ is preferred as it minimizes systematic uncertainties arising in the simulations. The ratio is given in Table 5.6 for the sum of internal and external coincidences. The ratio was found to vary around $\sim 0.22 - 0.27$ for external coincidences. Interestingly, even though no addback was utilized for internal coincidences, the three $\gamma$-ray decay can still be observed in the internal coincidence ROI due to coincident summing. This contribution is less probable and, as expected, has a ratio of $\sim 0.10 - 0.12$.

The improvement due to this effect depends on the branching ratio of the decay. As such, the correction was not included in the above efficiency values. When calculating limits on the half-life, the product of the branching ratio $f$ and the efficiency
Table 5.6: The contributions from the three-$\gamma$ decay mode into the two-$\gamma$ ROI.

<table>
<thead>
<tr>
<th>$J^\pi$</th>
<th>$\gamma_1$ [keV]</th>
<th>$\gamma_2$ [keV]</th>
<th>$\gamma_3$ [keV]</th>
<th>$\epsilon_{3\gamma}/\epsilon_{2\gamma}$</th>
<th>Run #1</th>
<th>Run #2</th>
</tr>
</thead>
<tbody>
<tr>
<td>0$^-$</td>
<td>709.9</td>
<td>1153.5</td>
<td>88.97</td>
<td>0.228 ± 0.003</td>
<td>0.190 ± 0.003</td>
<td></td>
</tr>
<tr>
<td>2$^+$</td>
<td>761.3</td>
<td>1153.5</td>
<td>88.97</td>
<td>0.226 ± 0.003</td>
<td>0.192 ± 0.003</td>
<td></td>
</tr>
<tr>
<td>2$^+$</td>
<td>849.6</td>
<td>1065.2</td>
<td>88.97</td>
<td>0.252 ± 0.004</td>
<td>0.212 ± 0.003</td>
<td></td>
</tr>
</tbody>
</table>

$\epsilon$ is summed over all decay modes. Assuming two possible decay modes, the effect is rewritten

$$
\sum_{n=2\gamma,3\gamma} f_n \epsilon_n = \epsilon_{2\gamma} \left( f_{2\gamma} + f_{3\gamma} \frac{\epsilon_{3\gamma}}{\epsilon_{2\gamma}} \right). \quad (5.5)
$$

This factor is used to calculate the half-life limits presented in the following section. In all cases, the branching ratio is $\approx 50\%$ for both the two and three $\gamma$-ray decay modes. As such, the ratio approximately corresponds to the sensitivity increase. An additional 6.0\% systematic uncertainty is assigned to the efficiency ratio to account for the differences between the GEANT4 simulation and the measured efficiency. Incorporating these effects, the effective branching ratio from the 1952 keV state increases from 44.7\% to 53.1\%. For the two decays from the 2003 keV state that exhibit ternary decays, via the 1242 and 1154 keV states, the effective branching ratio is 16.3\% and 7.35\%, respectively. This is an increase over the naive branching ratios of 13.7\% and 6.06\%, respectively. These numbers are for run #2, and are slightly larger for run #1.
Conclusion

In this chapter, the final results of all decay searches are presented. As no counts above background were observed, lower limits are calculated. For $\beta\beta$ decay, NMEs are extracted and the values are compared to theory.

6.1 Limit setting

Lower limits were set using the formula

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

where $N_0$ is the number of nuclei, $t$ is the exposure time, $f_b$ is the branching ratio, $\epsilon_{\gamma\gamma}^{tot}$ is the efficiency with all the corrections discussed in the previous chapters, 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 [86] for a Poisson process and a 90% confidence level. The 90% systematic uncertainty is included in limit setting, further reducing the limit to produce a more conservative result.
6.2 $\beta\beta$ decays of $^{96}\text{Zr}$ to excited final states

The $\beta\beta$ decay of $^{96}\text{Zr}$ to a number of different states in $^{96}\text{Mo}$ was studied. As the decay’s electrons are not detected, the limit is for the sum of the decay modes $2\nu\beta\beta + 0\nu\beta\beta$. Given present experimental limits, the $2\nu$ mode is of course more probable and the limits are effectively on the $2\nu$ mode. The enriched $^{96}\text{Zr}$ sample was counted for 1.922 yr. The number of nuclei is given by the masses in Sec. 3.5.1 divided by 96 g and multiplied by Avogadro’s number. As the $^{96}\text{Zr}$ sample consists of two samples with different radii, Eq. (6.1) is factorized such that the variables that depend on radius, $N_0$ and $\epsilon_{\gamma\gamma}^{\text{tot}}$, are grouped together, as shown in Eq. (4.8). The efficiency is calculated as shown in Sec. 4.4. The branching ratio, results of the sensitivity study from Sec. 4.3, and the systematic uncertainty are summarized in Table 6.1 for the different transitions. Two different statistical factors are given: $N_d$ and $N_s$. $N_d$ is the calculated 90% Feldman-Cousins upper confidence limit using the number of counts found in the ROI and the expected background. $N_s$ is the 90% Feldman-Cousins sensitivity. The sensitivity is only a function of the expected background and is the mean limit that would be measured by a collection of experiments with no signal. The confidence limit should be used in most cases. If the number of observed counts is less than the expected background, the confidence upper limit will be lower than an identical experiment that observes the exact number of expected background events. In this case, the Feldman-Cousins sensitivity is given in addition to the confidence limit. For the decay to the $0_1^+$ state, the most stringent confidence limit was produced by using the $\pm 3\sigma$ energy cut, while the most stringent sensitivity was produced by using the $\pm 2\sigma$ energy cut. The results of both cuts are shown in Table 6.1, with the best values being used to produce the final limits.

The limits for the half-life of $^{96}\text{Zr}$’s $\beta\beta$ decay to excited states of $^{96}\text{Mo}$ are given in Table 6.2. The same table also contains the previous best limits and theoretical
Table 6.1: For each of the excited states possible for the $\beta\beta$ decay of $^{96}\text{Zr}$, the region of interest, the number of observed events $n_{\text{obs}}$, the expected background $n_{\text{bkgd}}$, the Feldman-Cousins upper limit $N_d$, the Feldman-Cousins sensitivity $N_s$, and the systematic uncertainty $\sigma_{\text{syst}}$ are given.

<table>
<thead>
<tr>
<th>$J^\pi$</th>
<th>$\gamma_1$</th>
<th>$\gamma_2$</th>
<th>$f_b$</th>
<th>$n_{\text{obs}}$</th>
<th>$n_{\text{bkgd}}$</th>
<th>$N_d$</th>
<th>$N_s$</th>
<th>$\sigma_{\text{syst}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$0^+_1$</td>
<td>369.7</td>
<td>778.2</td>
<td>1.00</td>
<td>4*</td>
<td>5.34*</td>
<td>3.32*</td>
<td>5.29*</td>
<td>5.0%</td>
</tr>
<tr>
<td>$0^+_2$</td>
<td>369.7</td>
<td>778.2</td>
<td>1.00</td>
<td>2†</td>
<td>2.01†</td>
<td>3.90†</td>
<td>3.93†</td>
<td>5.0%</td>
</tr>
<tr>
<td>$0^+_3$</td>
<td>551.8</td>
<td>778.2</td>
<td>1.00</td>
<td>3</td>
<td>1.50</td>
<td>5.93</td>
<td>3.60</td>
<td>5.1%</td>
</tr>
<tr>
<td>$2^+_2$</td>
<td>719.6</td>
<td>778.2</td>
<td>0.703</td>
<td>2</td>
<td>0.97</td>
<td>4.94</td>
<td>3.20</td>
<td>5.2%</td>
</tr>
<tr>
<td>$2^+_3$</td>
<td>847.7</td>
<td>778.2</td>
<td>0.903</td>
<td>2</td>
<td>0.87</td>
<td>5.04</td>
<td>2.97</td>
<td>5.3%</td>
</tr>
<tr>
<td>$0^+_3$</td>
<td>1844.3</td>
<td>778.2</td>
<td>1.00</td>
<td>1</td>
<td>1.27</td>
<td>3.11</td>
<td>3.47</td>
<td>5.7%</td>
</tr>
</tbody>
</table>

* Using the $\pm 3\sigma$ cut.
† Using the $\pm 2\sigma$ cut.

Calculations for the decay. It can be seen that the current limits are an improvement over the previous best work by a factor of five for the $0^+_1$ state and a factor of 2 for the $2^+_2$ and $2^+_3$ states. Further improvements would necessitate increasing the $^{96}\text{Zr}$ sample size or a customized detector with higher efficiency.

A systematic law for the $\beta\beta$-decay half-lives, published by Ren and Ren [37], is used to produce estimates for the excited state decays. This law is fit to experimental data for the ground state decay, but was shown to produce accurate results for decays to the first excited $0^+$ state. The law agreed within the experimental error for the decay of $^{100}\text{Mo}$ to the $0^+_1$ state. The law underestimated the half-life of $^{150}\text{Nd}$ by 30-70%, although the experimental errors are large and the deformation of $^{150}\text{Nd}$ is not taken into account. It is of some interest then, that our experimental limit exceeds the prediction of Ref. [37] by 25%. The present work excludes this prediction at the 93% confidence level. This law is also used to calculate estimates for decays to the excited $2^+$ states, but it must be noted that this law was not calibrated for decays to $2^+$ states and may not produce reliable results. Two theoretical estimates using
Table 6.2: Half-life limits at the 90% confidence level on the $\beta\beta$ decay of $^{96}$Zr to different excited states are given. As a comparison, the previous best limits and theoretical results are also shown. Note that the method of Ref. [37] is not calibrated for decays to $2^+$ states and may not produce reliable results for these decay modes.

<table>
<thead>
<tr>
<th>$J^+$</th>
<th>$E$ [keV]</th>
<th>$T_{1/2}$ [yr] This work</th>
<th>$T_{1/2}$ [yr] This work C.L. Sensitivity</th>
<th>$T_{1/2}$ [yr] Previous work</th>
<th>$T_{1/2}$ [yr] Systematic law</th>
<th>$T_{1/2}$ [yr] Theory</th>
</tr>
</thead>
<tbody>
<tr>
<td>$0^+_1$</td>
<td>1148.1</td>
<td>$&gt;3.2 \times 10^{20}$</td>
<td>$&gt;2.8 \times 10^{20}$</td>
<td>$&gt;6.8 \times 10^{19}$</td>
<td>$2.59 \times 10^{20}$</td>
<td>$3.8 \times 10^{21}$</td>
</tr>
<tr>
<td>$0^+_2$</td>
<td>1330.0</td>
<td>$&gt;1.4 \times 10^{20}$</td>
<td>-</td>
<td>-</td>
<td>$4.29 \times 10^{20}$</td>
<td>-</td>
</tr>
<tr>
<td>$2^+_2$</td>
<td>1497.8</td>
<td>$&gt;1.0 \times 10^{20}$</td>
<td>-</td>
<td>$&gt;6.1 \times 10^{19}$</td>
<td>$7.42 \times 10^{20}$</td>
<td>-</td>
</tr>
<tr>
<td>$2^+_3$</td>
<td>1625.9</td>
<td>$&gt;1.2 \times 10^{20}$</td>
<td>-</td>
<td>$&gt;5.4 \times 10^{19}$</td>
<td>$1.21 \times 10^{21}$</td>
<td>-</td>
</tr>
<tr>
<td>$0^+_3$</td>
<td>2622.5</td>
<td>$&gt;1.1 \times 10^{20}$</td>
<td>$&gt;1.0 \times 10^{20}$</td>
<td>-</td>
<td>$2.08 \times 10^{25}$</td>
<td>-</td>
</tr>
</tbody>
</table>
microscopic models are also given for the decay. Both are older works, published in 1996, and as such do not include recent advancements made in 2νββ-decay NME theory. The calculations of Ref. [89] use QRPA techniques, while Ref. [90] uses the RQRPA model. A comparison to more recent work is done by extracting a limit on the NME.

A limit on the decay’s NME may be extracted using Eq. (2.22) and the phase-space factor given in Table 1.3. The phase-space factor has only been calculated for the decay to the first excited 0^+_1 state. This results in a limit on the NME |M_2νββ(0^+_1)| < 0.11 at the 90% confidence level. Unfortunately, this is not strict enough to test the calculation of Ref. [51], who calculated |M_2νββ(0^+_1)| = 0.04. Their reported calculation was done using the closure approximation, which is often disfavored for the SSD hypothesis. It would be interesting to see how this limit compared to a calculation performed using the SSD hypothesis.

6.3 β decay of ⁹⁶Zr

Limits for the β decay of ⁹⁶Zr were also extracted from the same data set. The analysis and efficiency calculations were detailed in Chapter 4. The three most probable decay sequences were investigated, in total representing 78.9% of all β decays. Each decay sequence produces three coincidence γ rays, which leads to six coincident pairs in the detectors, as was outlined in Chapter 4. Three of these ROIs are not included due to background from naturally occurring radiation. The individual results for each of these coincidence pairs are given in Table 6.3. The Feldman-Cousins confidence interval construction is used and all results are reported at the standard 90% confidence level.

The results of these searches may be combined to form one statistically significant result. The individual results are summed before the limit is extracted. Once again, the Feldman-Cousins method is used. Here, the number of observed events
Table 6.3: Half-life limits for $^{96}$Zr’s $\beta$ decay are summarized. For each decay sequence and coincident pair the Feldman Cousins 90% confidence limit and sensitivity are both given. The final half-life lower limit is calculated using the confidence limit.

<table>
<thead>
<tr>
<th>Decay sequence</th>
<th>Given $\gamma$ $[\text{keV}]$</th>
<th>Second $\gamma$ $[\text{keV}]$</th>
<th>$N_d$</th>
<th>$N_s$</th>
<th>$T_{1/2}^{1/2}$ $[\text{yr}]$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$5^+_0 \rightarrow 4^+_1 \rightarrow 2^+_0 \rightarrow 0^+_0$</td>
<td>$1091.3$</td>
<td>$568.9$</td>
<td>$6.08$</td>
<td>$3.53$</td>
<td>$&gt; 9.5 \times 10^{18}$</td>
</tr>
<tr>
<td></td>
<td>$1091.3$</td>
<td>$778.2$</td>
<td>$3.27$</td>
<td>$3.35$</td>
<td>$&gt; 1.3 \times 10^{19}$</td>
</tr>
<tr>
<td></td>
<td>$1091.3$</td>
<td>$1347.1^*$</td>
<td>$2.20$</td>
<td>$2.65$</td>
<td>$&gt; 3.8 \times 10^{18}$</td>
</tr>
<tr>
<td></td>
<td>$568.9$</td>
<td>$1869.5^*$</td>
<td>$2.08$</td>
<td>$2.75$</td>
<td>$&gt; 4.5 \times 10^{18}$</td>
</tr>
<tr>
<td></td>
<td>$778.2$</td>
<td>$1660.2^*$</td>
<td>$2.26$</td>
<td>$2.60$</td>
<td>$&gt; 4.2 \times 10^{18}$</td>
</tr>
<tr>
<td>$5^+_0 \rightarrow 3^+_0 \rightarrow 2^+_0 \rightarrow 0^+_0$</td>
<td>$460.0$</td>
<td>$1200.0$</td>
<td>$2.54$</td>
<td>$3.91$</td>
<td>$&gt; 9.1 \times 10^{18}$</td>
</tr>
<tr>
<td></td>
<td>$778.2$</td>
<td>$1200.0$</td>
<td>$3.59$</td>
<td>$3.1$</td>
<td>$&gt; 4.1 \times 10^{18}$</td>
</tr>
<tr>
<td></td>
<td>$778.2$</td>
<td>$1660.0^*$</td>
<td>$2.29$</td>
<td>$2.57$</td>
<td>$&gt; 1.6 \times 10^{18}$</td>
</tr>
<tr>
<td></td>
<td>$1200.0$</td>
<td>$1238.2^*$</td>
<td>$4.09$</td>
<td>$2.68$</td>
<td>$&gt; 8.2 \times 10^{17}$</td>
</tr>
<tr>
<td>$5^+_0 \rightarrow 4^+_0 \rightarrow 2^+_0 \rightarrow 0^+_0$</td>
<td>$778.2$</td>
<td>$849.9$</td>
<td>$4.51$</td>
<td>$3.56$</td>
<td>$&gt; 2.7 \times 10^{18}$</td>
</tr>
<tr>
<td></td>
<td>$778.2$</td>
<td>$810.8$</td>
<td>$5.86$</td>
<td>$3.67$</td>
<td>$&gt; 1.9 \times 10^{18}$</td>
</tr>
<tr>
<td></td>
<td>$849.9$</td>
<td>$810.8$</td>
<td>$1.47$</td>
<td>$3.46$</td>
<td>$&gt; 8.0 \times 10^{18}$</td>
</tr>
<tr>
<td></td>
<td>$778.2$</td>
<td>$1660.7^*$</td>
<td>$2.23$</td>
<td>$2.63$</td>
<td>$&gt; 8.6 \times 10^{17}$</td>
</tr>
<tr>
<td></td>
<td>$849.9$</td>
<td>$1589.0^*$</td>
<td>$2.35$</td>
<td>$2.52$</td>
<td>$&gt; 7.5 \times 10^{17}$</td>
</tr>
<tr>
<td></td>
<td>$810.8$</td>
<td>$1628.1^*$</td>
<td>$4.23$</td>
<td>$2.56$</td>
<td>$&gt; 4.5 \times 10^{17}$</td>
</tr>
</tbody>
</table>

* Sum of two coincident $\gamma$ rays

exceeds the background, so only the Feldman-Cousins limit is reported, not the sensitivity. This analysis is shown in Table 6.4. The final result is a combined limit of $T_{1/2}^{1/2}(\beta(^{96}\text{Zr})) > 2.4 \times 10^{19}$ 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. This is because additional transitions increase the background, thus reducing the detectors total sensitivity, at the same rate at which they increase the total efficiency. The limit produced using only the search for two $\gamma$-ray events is a very comparable $2.1 \times 10^{19}$ yr. This same statistical analysis was also performed using the Bayesian approach of Ref. [91]. A step function was used as the prior and the posterior was proportional to the product of the independent likelihood functions for each individual ROI. This technique was found to produce a combined limit of $T_{1/2} > 2.0 \times 10^{19}$ yr at the 90% confidence level. This approach was abandoned in favored of the more stringent limit produced.
Table 6.4: Half-life limits for $^{96}\text{Zr}$’s $\beta$ decay from the combined statistical analysis at the 90% confidence level.

<table>
<thead>
<tr>
<th>Decay sequence</th>
<th>Two-$\gamma$ events [yr]</th>
<th>Three-$\gamma$ events [yr]</th>
<th>Combined [yr]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$5_0^+ \rightarrow 4_1^+ \rightarrow 2_0^+ \rightarrow 0_0^+$</td>
<td>$1.6 \times 10^{19}$</td>
<td>$1.6 \times 10^{19}$</td>
<td>$2.4 \times 10^{19}$</td>
</tr>
<tr>
<td>$5_0^+ \rightarrow 3_0^+ \rightarrow 2_0^+ \rightarrow 0_0^+$</td>
<td>$1.1 \times 10^{19}$</td>
<td>$0.18 \times 10^{19}$</td>
<td>$1.0 \times 10^{19}$</td>
</tr>
<tr>
<td>$5_0^+ \rightarrow 4_0^+ \rightarrow 2_0^+ \rightarrow 0_0^+$</td>
<td>$0.61 \times 10^{19}$</td>
<td>$0.14 \times 10^{19}$</td>
<td>$0.66 \times 10^{19}$</td>
</tr>
<tr>
<td>Combined</td>
<td>$2.1 \times 10^{19}$</td>
<td>$0.94 \times 10^{19}$</td>
<td>$2.4 \times 10^{19}$</td>
</tr>
</tbody>
</table>

from summing the individual results.

Unfortunately, this limit is not sufficient to test the theoretical limit of $2.4 \times 10^{20}$ yr by Ref. [50]. Furthermore, the present limit is less than the previous limit of $3.8 \times 10^{19}$ [83]. The previous work used a single detector to search for the 778.2 keV $\gamma$ ray. This results in a higher efficiency, but with an increased background. As the present experiment covers a larger solid angle, the coincidence summing of the three coincident $\gamma$ rays becomes a large factor and noticeably decreases the efficiency compared to a single detector with half the solid angle. As has been previously stated, although the technique of Ref. [83] is sufficient for limit setting, any observation would require detection of coincident $\gamma$ rays to distinguish the $\beta$ and $\beta\beta$ decay to excited states. The background reduction provided by the $\gamma - \gamma$ coincidence has also been shown in the present work and will be necessary in any future searches. As such, the present experiment has a much higher discovery potential even though the produced limits may not be as competitive.

6.4 $ECEC$ decays of $^{156}\text{Dy}$

Four states were identified as possibilities for resonant-$0\nu ECEC$ transitions in Ref. [44]. Each of these states has a distinct ROI that was analyzed separately by the procedures in Chapter 5. The results of these analyses are summarized in Table 6.5. These results are for the sum of run #1 and run #2, which produces better limits than ei-
Table 6.5: For each $ECEC$ decay mode studied, the ROI, effective branching ratio $f_{b,\text{eff}}$, number of observed events $n_{\text{obs}}$, number of background events $n_{\text{bkgd}}$, the Feldman-Cousins upper limit $N_d$, the Feldman-Cousins sensitivity $N_s$, and the systematic uncertainty $\sigma_{\text{syst}}$ are given.

<table>
<thead>
<tr>
<th>$J^\pi$</th>
<th>$\gamma_1$ [keV]</th>
<th>$\gamma_2$ [keV]</th>
<th>$f_{b,\text{eff}}$</th>
<th>$n_{\text{obs}}$</th>
<th>$n_{\text{bkgd}}$</th>
<th>$N_d$</th>
<th>$N_s$</th>
<th>$\sigma_{\text{syst}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$1^-$</td>
<td>1857.4</td>
<td>88.97</td>
<td>0.578</td>
<td>1</td>
<td>4.12</td>
<td>1.28</td>
<td>4.86</td>
<td>5.6%</td>
</tr>
<tr>
<td>$0^-$</td>
<td>709.9</td>
<td>1242.5</td>
<td>0.531</td>
<td>2</td>
<td>2.38</td>
<td>3.55</td>
<td>4.12</td>
<td>5.7%</td>
</tr>
<tr>
<td>$0^+_4$</td>
<td>1899.5</td>
<td>88.97</td>
<td>1*</td>
<td>2</td>
<td>3.76</td>
<td>2.49</td>
<td>4.72</td>
<td>5.6%</td>
</tr>
<tr>
<td>$2^+$</td>
<td>684.0</td>
<td>1319.7</td>
<td>0.061</td>
<td>1</td>
<td>1.92</td>
<td>2.59</td>
<td>3.87</td>
<td>5.7%</td>
</tr>
<tr>
<td>$2^+$</td>
<td>761.3</td>
<td>1242.5</td>
<td>0.163</td>
<td>4</td>
<td>2.74</td>
<td>5.86</td>
<td>4.29</td>
<td>5.7%</td>
</tr>
<tr>
<td>$2^+$</td>
<td>849.6</td>
<td>1154.2</td>
<td>0.074</td>
<td>4</td>
<td>2.62</td>
<td>5.99</td>
<td>4.23</td>
<td>5.7%</td>
</tr>
<tr>
<td>$2^+$</td>
<td>849.6</td>
<td>1154.2</td>
<td>0.076†</td>
<td>1†</td>
<td>0.99†</td>
<td>3.37†</td>
<td>3.27†</td>
<td>5.7%</td>
</tr>
</tbody>
</table>

* As this branching ratio has not been measured, it is assumed to be 1.
† Only external coincidences were used in this analysis.

ther run alone. This represents a combined run time of 0.635 yr and a total exposure of 0.119 g·yr of $^{156}$Dy. The Feldman-Cousins upper limit and sensitivity are both given at the 90% confidence level. As the $2^+$ 2003.7 keV state has three separate two $\gamma$-ray transitions with a significant branching ratio, the results are listed for all three. For the 849.6+1154.2 keV coincidence, the background rate was found to be sufficiently high that a better limit could be obtained by only including external coincidences. This lowers both the efficiency and the Feldman-Cousins upper limit $N_d$. The results for both analysis procedures are given in Table 6.5 in order to show the improvement.

As no statistically significant counts were observed above background, limits for resonant $0\nu ECEC$ were set and are given in Table 6.6. In calculating these limits, it was assumed that the $0^+_4$ state decays entirely through the $2^+_5$ state, as discussed in Sec. 5.3.3. These limits may be easily updated, should the branching ratio be measured, by simply multiplying the limit by the measured branching ratio. The three decay modes from the 2003.7 keV state are summed together to present a single confidence limit. The results of this procedure are shown in Table 6.7.
Table 6.6: The final half-life limits for the ECEC decay of $^{156}$Dy to excited states at the 90% confidence level. Both the confidence limit, the experimental sensitivity, and the previous measurement are given.

<table>
<thead>
<tr>
<th>$J^\pi$</th>
<th>$E$ [keV]</th>
<th>Lim $T_{1/2}$ [yr] This work</th>
<th>Lim $T_{1/2}$ [yr] This work</th>
<th>Lim $T_{1/2}$ [yr] Previous limit</th>
</tr>
</thead>
<tbody>
<tr>
<td>1$^-$</td>
<td>1946.4</td>
<td>$1.0 \times 10^{18}$</td>
<td>$2.8 \times 10^{17}$</td>
<td>$9.6 \times 10^{15}$</td>
</tr>
<tr>
<td>0$^-$</td>
<td>1952.4</td>
<td>$2.2 \times 10^{17}$</td>
<td>$1.9 \times 10^{17}$</td>
<td>$2.6 \times 10^{16}$</td>
</tr>
<tr>
<td>0$^+_4$</td>
<td>1988.5</td>
<td>$9.5 \times 10^{17*}$</td>
<td>$5.0 \times 10^{17*}$</td>
<td>$1.9 \times 10^{16}$</td>
</tr>
<tr>
<td>2$^+$</td>
<td>2003.7</td>
<td>$6.7 \times 10^{16}$</td>
<td>-</td>
<td>$3.0 \times 10^{14}$</td>
</tr>
</tbody>
</table>

* This limit is calculated for the $0^+_4 \rightarrow 2^+ \rightarrow 0^+_0$ decay mode and assumes a branching ratio of 1. If the branching ratio is measured to be less than one, these values must be multiplied by the new branching ratio.

Table 6.7: The half-life limits for the ECEC decay of $^{156}$Dy to the 2003.7 keV state at the 90% confidence level. The final limit is produced from the combined statistics of the three studied decay modes.

<table>
<thead>
<tr>
<th>Decay sequence</th>
<th>Lim $T_{1/2}$ [yr]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$2^+ \rightarrow 2^- \rightarrow 0^+_0$</td>
<td>$3.4 \times 10^{16}$</td>
</tr>
<tr>
<td>$2^+ \rightarrow 1^- \rightarrow 0^+_0$</td>
<td>$3.9 \times 10^{16}$</td>
</tr>
<tr>
<td>$2^+ \rightarrow 2^+ \rightarrow 0^+_0$</td>
<td>$2.2 \times 10^{16}$</td>
</tr>
<tr>
<td>Combined</td>
<td>$6.7 \times 10^{16}$</td>
</tr>
</tbody>
</table>

the decay of the 2003.7 keV state through the 1154.2 keV 2$^+$ state, only external coincidences were used in order to produce the most stringent limit. If both internal and external coincidences are included, the combined lower limit for the decay from the 2003.7 keV state is lowered to $5.6 \times 10^{16}$ yr. The number of counts in the ROI exceeds the expected background for this transition, so the half-life limit is lower than the expected sensitivity. As with the previous section, the experimental sensitivity is only given for decays in which it is lower than the set limit.

The limits produced by the current work represent a large improvement over the previous best limits measured by Ref. [85]. The limit to the $1^-$ state is improved by a factor of 29, the $0^-$ state by a factor of 7.3, the $0^+_4$ state by a factor of 26, and the $2^+$ state by a factor of 220. This is much in part due to the higher efficiency of the
two-clover apparatus and use of an isotopically enriched sample. The current limits are not able to provide any competitive limits on the neutrino mass or the $ECEC$ matrix elements. A much larger sample mass is necessary in order to test the theory of Majorana neutrinos using resonant $0\nu ECEC$. $^{156}$Dy samples are of course limited by the small natural abundance of this isotope: 0.056%.

Using the presented results, one can envision designing a large-scale $ECEC$ experiment. A large-scale experiment would necessitate enriched samples, which the present work shows can be produced without large radioactive backgrounds in the region of interest. The experiment would also greatly benefit from the background reduction provided by detection of the individual $\gamma$ rays using the coincidence technique. This would be best accomplished using a segmented detector, as was done here. Additional improvements could be made by using pulse shape discrimination to perform a $\gamma$-ray tracking within each segment. This would help distinguish Compton-scattered $\gamma$ rays from signal events. Although the energy resolution of HPGe detectors is essential in the present work, a large-scale experiment would want to investigate a detector material containing Dy. By making the detector out of the source material, the flexibility of changing the source is lost but it becomes much easier to scale the experiment to a larger size. A large-scale experiment, however, is not necessary until a viable resonant $0\nu ECEC$ candidate isotope is identified. The present two-clover apparatus has been proven to produce high-quality $ECEC$ half-life limits for $^{156}$Dy with the ability to measure any candidate isotopes that may emerge with new precision mass measurements.

6.5 Concluding remarks

Numerous $0\nu\beta\beta$ decay experiments are currently underway with plans to progress towards a ton-scale experiment in the near future. These experiments may soon illuminate the Majorana or Dirac nature of the neutrino. If a measurement of $0\nu\beta\beta$
is made, accurate NME calculations are necessary to extract the neutrino mass. Data on the $2\nu\beta\beta$ decay, such as on the decay to excited states provided here, are necessary to test and study $\beta\beta$ decay NMEs. The case of $^{96}$Zr, studied herein, is of interest because it has the third largest phase space for the decay to the first excited $0^+_1$ state of all candidate nuclei. Although a suppression of the NME to the $0^+_1$ state predicted by Ref. [51] is somewhat confirmed, $^{96}$Zr remains an intriguing candidate for nuclear structure effects on the $2\nu\beta\beta$ decay NME thanks two its closed shell structure. The limits on the $\beta\beta$ decay to excited states presented are currently the most competitive limits for $^{96}$Zr.

Resonant $0\nu ECEC$ to an excited state is a valid alternative to $0\nu\beta\beta$, should a suitable candidate nucleus be discovered. It is then important to perform experiments on candidate nuclei to explore this alternative. The half-life limits measured in this thesis are the most stringent limits for $0\nu ECEC$ in $^{156}$Dy. With hope, the two second-order weak decays studied in this thesis will one day provide breakthroughs in our knowledge and understanding of the neutrino and weak interaction.
Bibliography


Biography

Sean William Finch was born September 11, 1987, in Wilson, NC. He received Bachelor of Science degrees in both Physics and Applied Mathematics from North Carolina State University in 2009. In 2013 he received his Master of Arts from Duke University in Physics. He continued to graduate from Duke University in 2015 with his PhD in physics.

While studying at Duke University, Sean received the departmental Outstanding Teaching Assistant Award in 2010 for his work teaching undergraduate labs. In 2012 Sean received the Henry W. Newson Graduate Fellowship for his contributions as a graduate researcher.

Publications

1. M. F. Kidd, J. H. Esterline, S. W. Finch, and W. Tornow, Two-neutrino double-$\beta$ decay of $^{150}$Nd to excited final states in $^{150}$Sm, Phys. Rev. C 90, 055501 (2014)


3. C. Bhatia, S. W. Finch, M. E. Gooden, and W. Tornow, $^{40}$Ar($n, p)^{40}$Cl reaction cross section between 9 and 15 MeV, Phys. Rev. C 86, 041602(R) (2012)