Geochemical measurement of the half-life of the double-beta decay of $^{96}\text{Zr}$

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Project Overview

- Studying the decay ($\beta\beta$ and $\beta$) of $^{96}\text{Zr} \rightarrow ^{96}\text{Mo}$
  - System provides unique insights into double-beta decay
- We are studying two properties:
  - Q value (completed 2015, published PRL 116 (2016))
  - Half-life (completed 2017, publishing soon)
\[96\text{Zr} \text{ Decay}\]

- \(96\text{Zr}\) is of particular interest:
  - One of the largest Q values and shortest half-lives
  - Unstable against single \(\beta\)-decay (4\(^\text{th}\) order forbidden)
- Previous measurements of half-life did not agree:
  - Geochemical measurements:
    - \(3.9(9) \times 10^{19}\) yr (1993)
    - \(0.94(32) \times 10^{19}\) yr (2001)
  - Direct count-rate measurement:
    - \(2.35(30) \times 10^{19}\) yr (2010)
- Zircon, or ZrSiO$_4$, is a highly stable mineral
- Can remain a closed system over billions of years
  - Evidenced by accurate (concordant) U-Pb ages
- Large amount of zirconium (~50 wt%)
  - $^{96}$Zr is 2.80% of zirconium
- Very little molybdenum (< ppm)
Double-beta decay half-life by stable isotope geochemistry

Isotopic Composition of Mo

Zircon

0.5 – 2.5 Ga
ZrSiO₄

2.8 %^{96}Zr
^{96}Zr \rightarrow ^{96}Mo + 2e^- + 2\nu

\begin{align*}
  ^{96}Zr & \rightarrow ^{96}Mo \\
  \text{Isotopic Abundance} & \\
  \text{Mass Number} & \\
  92 & 93 & 94 & 95 & 96 & 97 & 98 & 99 & 100
\end{align*}
Zircon sample prep and analysis

- 10 mL HF
- 500 mg zircon
- $220^\circ C$

**Ion Exchange**

**Thermo Scientific Neptune:**
Multi-collector inductively coupled plasma mass spectrometer

**Need maximum recovery**
(achieved $>60\%$)

**Need $>10^7$ reduction in Zr**
(achieved $\sim 10^9$ !)

**Zr and Ru isobaric interference correction;**
$\leq 50$ ppm precision
(achieved 20 ppm)
Half-life calculation

\[ t_{1/2} = \frac{-t \ln(2)}{\ln(1 - n_d/n_0)} \]

\( n_d \) daughter product:
\[ n_d(^{96}\text{Mo}) = \frac{m_{\text{Mo}} N_A}{A_W(\text{Mo})} C(^{96}\text{Mo}) \delta(^{96}\text{Mo}) \]

\( n_0 \) parent:
\[ n_0(^{96}\text{Zr}) \cong \frac{m_{\text{Zr}} N_A}{A_W(\text{Zr})} C(^{96}\text{Zr}) \]

\[ \frac{n_d}{n_0} = \frac{m_{\text{Mo}}}{m_{\text{Zr}}} \frac{A_W(\text{Zr})}{A_W(\text{Mo})} \frac{C(^{96}\text{Mo})}{C(^{96}\text{Zr})} \delta(^{96}\text{Mo}) \]
Geochemical results

- Red shows 2.68 Gyr zircon
- Stars show expected U spontaneous fission decay
- Black shows 1 Gyr zircon

$^{97}\text{Mo}/^{95}\text{Mo fixed to expected U-SF decay pattern}$
\[
T_{1/2} = 2.03^{+0.46}_{-0.31} \cdot 10^{19} \text{ yr}
\]

Compare this to NEMO $\beta\beta$ half-life of: 
\[
T_{1/2}^{\beta\beta} (2.35 \pm 0.21) \cdot 10^{19} \text{ yr}
\]

\[
T^\beta_{1/2} > 5.2 \cdot 10^{19} \text{ yr}
\]
## Contributions to uncertainty

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Contribution</th>
</tr>
</thead>
<tbody>
<tr>
<td>Age</td>
<td>0.0%</td>
</tr>
<tr>
<td>$[U]$</td>
<td>35.0%</td>
</tr>
<tr>
<td>mMo/mZr</td>
<td>6.2%</td>
</tr>
<tr>
<td>Zr Composition</td>
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</tr>
<tr>
<td>Mo Composition</td>
<td>7.1%</td>
</tr>
<tr>
<td>Delta-96Mo</td>
<td>49.0%</td>
</tr>
</tbody>
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Improvements to results require better $\delta^{(96}\text{Mo})$ result. To achieve this would require a larger sample with a lower “common” Mo content.
Future endeavors

- We have resolved the discrepancy between previous geochemical measurements and counting-rate experiments.
- Search for new high-quality ancient zircon samples with lower Mo content to measure.
  - A significant improvement in uncertainty is possible, and could yield the first measurement of the $^{96}$Zr single-beta decay rate.
- Develop technique to fully separate ruthenium from very large quantity of zirconium ($>10^{10}$ separation required).
  - Possible to improve lower limits of $^{96}$Zr quadruple-beta decay rate by **two orders of magnitude**.