Isotope abundance measurement of the half-life of the $\beta\beta$-decaying nucleus ^{96}Zr from a 2.68 Gyr zircon sample

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The decay half-life of the ^{96}Zr isotope was measured by applying isotope geochemistry techniques to ancient (0.9 and 2.68 Gyr) $ZrSiO_4$ (zircon) samples, with the objective to separate the single and double β-decay branches. The single β decay provides one of the most direct tests for theoretical models describing neutrinoless $\beta\beta$ decays. Both the single and the double β-decay branches lead to the same final nucleus ^{96}Mo and generate an isotopic anomaly over geological time scales. After a chemical separation, the Mo isotopic composition was measured by inductively coupled plasma mass spectrometry (ICPMS). The ^{96}Mo isotopic anomaly was determined in two complete replicate analyses to be 107(40) and 88(20) ppm, which translates to a ^{96}Zr half-life of $T_{1/2} = (2.03^{+0.46}_{-0.31}) \times 10^{19}$ yr. With the $2\nu\beta\beta$ partial decay half-life of the ground-state to ground-state transition known from NEMO-3 to be $2.35 \pm 0.21 \times 10^{19}$ yr and all other partial $\beta\beta$-decay half-lives expected to be many orders of magnitude longer, a lower limit for the single-β-decay half-life is set at $T^{\beta}_{1/2} \geqslant 6.2 \times 10^{19}$ yr.

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I. INTRODUCTION

Double-beta ($\beta\beta$) decay measurements are a class of nuclear studies with the objective of detecting the neutrinoless (0ν) decay variants. An observation of this decay mode provides insight into the nature and absolute mass scale of the neutrino. Critical to the theoretical descriptions of $0\nu\beta\beta$ decay are the nuclear matrix elements, which describe the underlying nuclear physics [1–3]. The nucleus ^{96}Zr is of particular interest as it is also unstable against single β decay, a property shared only with ^{48}Ca, and among these two it is experimentally the most advantageous because of its higher isotopic abundance and its shorter lifetime as predicted by theory [4–6].

The main branch of the ^{96}Zr single β decay proceeds through the $^{96}\mathrm{Zr}(J^{\pi} = 0^{+}) \rightarrow {}^{96}\mathrm{Nb}(44\ \mathrm{keV}, J^{\pi} = 5^{+})$ fourfold unique forbidden transition with a half-life estimated to be of the same order of magnitude as the well-known $2\nu\beta\beta$ decay [7,8]. Further, the uniqueness of the transition ensures that there is only one matrix element active, thereby providing a particularly clean test of the underlying models, which are applied to evaluate the $0\nu\beta\beta$-decay nuclear matrix elements [4]. The decay scheme of ^{96}Zr and the energetics of the various decay modes are shown in Fig. 1.

To date, the single β-decay half-life for ^{96}Zr has not been determined experimentally and only lower bounds have been established. The most recent one is $T^{\beta}_{1/2} > 2.6 \times 10^{19}$ yr [9], which supersedes an earlier value given in Ref. [10]. Two theoretical calculations for the single β-decay half-life exist, with values of $T^{\beta}_{1/2} = (11 g_{\mathrm{A}}^{-2}) \times 10^{19}$ yr and $T^{\beta}_{1/2} = (24 g_{\mathrm{A}}^{-2}) \times 10^{19}$ yr [4,7]. The first is based on the shell model and the second on the quasiparticle random-phase approximation (QRPA) model. In both cases the axial-vector coupling constant (g_{A}) appears as a yet to be determined parameter, whose value will likely fall somewhere between ≈0.9 (quenched value) and 1.27 (free value). It is worthwhile to note that both models are used as well for calculating the $0\nu\beta\beta$ decay.

In the geological sciences the Zr-containing mineral zircon ($ZrSiO_4$) constitutes a well-known and rather unique system because its lattice structure may remain an entirely closed system over geological time scales. Zircon can be used as a reliable, high-precision geochronometer by applying the uranium-lead (U-Pb) dating technique [11]. This technique is possible because trace amounts of U are incorporated into the zircon lattice structure during the time of rock formation, whereas Pb is strongly rejected. Pb then accumulates over time as a result of the U-decay chains, and can be quantified to estimate the age of the host zircon. Similarly, the isotope ^{96}Zr constitutes 2.80% of the total Zr content, and Mo content within zircon is often less than 1 part-per-million (ppm). It is thus possible to measure the ^{96}Zr-decay product, ^{96}Mo, as an excess relative to the natural composition. The age, low Mo content and chemical inertness make ancient zircon minerals a suitable system to study long-lived nuclear processes such as the $\beta\beta$ decay of the isotope ^{96}Zr.

Two previous measurements of ^{96}Zr decay yielded significantly different results. A Mo-isotope abundance measurement of a 1.8 Gyr zircon sample by Wieser and DeLaeter found a half-life of $0.94(32) \times 10^{19}$ yr [12], while a direct $\beta\beta$-decay counting experiment by the NEMO collaboration found a

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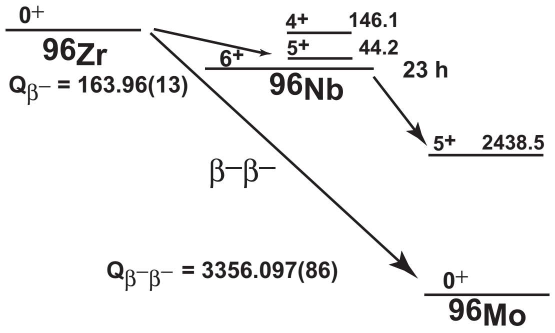

FIG. 1. Decay scheme of the $A = 96$ triplet showing the energy position of ^{96}Zr with respect to its neighbors ^{96}Nb and ^{96}Mo [4]. Indicated is the dominant single β-decay path from the ^{96}Zr ground state to the $J^\pi = 5^+$ state, and the ^{96}Nb ground state decays dominantly to the $J^\pi = 5^+$ excited state in ^{96}Mo.

half-life of $2.35(21) \times 10^{19}$ yr [13]. This has led to the speculation that there could be a contribution to the overall decay from the single β-decay of ^{96}Zr at a higher rate than is expected according to theory. It was therefore necessary to confirm the isotope abundance measurement with improved techniques [13].

II. DETAILS OF ISOTOPE ABUNDANCE MEASUREMENTS

An isotope abundance measurement of the ^{96}Zr decay half-life is performed by measuring the accumulated decay product ^{96}Mo as an excess relative to the natural isotopic composition after taking into account all known mass-dependent fractionation effects. The measured excess is used along with the ratio of Mo to Zr in the zircon sample to determine the relative amount of the daughter product according to the following equation:

$$\frac{n_d}{n_0} = \frac{m_{\rm Mo}}{m_{\rm Zr}} \frac{A_w({\rm Zr})}{A_w({\rm Mo})} \frac{C(^{96}{\rm Mo})}{C(^{96}{\rm Zr})} \delta(^{96}{\rm Mo}), \tag{1}$$

where n_d and n_0 are the amounts of daughter and parent, respectively; $m_{\rm Mo}$ and $m_{\rm Zr}$ are the total masses of Mo and Zr in the sample; $A_w({\rm Mo})$ and $A_w({\rm Zr})$ are the atomic weights of each element; $C(^{96}{\rm Mo})$ and $C(^{96}{\rm Zr})$ are the natural isotopic abundances of the respective isotopes; and $\delta(^{96}{\rm Mo})$ is the measured ^{96}Mo excess. The $\delta(^{96}{\rm Mo})$ value, referred to as a "δ-value," specifically represents the relative change in the $n(^{96}{\rm Mo})/n(^{95}{\rm Mo})$ isotope amount ratio, therefore any change in the amount of ^{95}Mo needs to be accounted for to extract the change in the amount of ^{96}Mo. The half-life $T_{1/2}$ is then determined from the n_d/n_0 ratio and the average age t of the zircon sample according to

$$T_{1/2} = \frac{-t \ln 2}{\ln{(1 - n_d/n_0)}}. \tag{2}$$

There are three values which have to be measured to determine the half-life: (i) the ^{96}Mo excess; (ii) the Mo:Zr mass ratio; and (iii) the age of the sample. The Mo isotopic composition can be measured by isotope-ratio mass spectrometry to determine the $\delta(^{96}{\rm Mo})$ value, and requires the sample to be dissolved and Mo be separated from Zr to eliminate isobaric interferences from Zr isotopes. The Mo:Zr mass ratio can similarly be measured with an isotope-ratio mass spectrometer. Lastly, the age is determined by U-Pb dating using isotope dilution thermal ionization mass spectrometry (ID-TIMS).

The previous geochemical experiment reported in Ref. [12] relied on the measurement of excesses of the Mo isotopes of mass 95, 97, 98, and 100 generated by the spontaneous fission of U to estimate the Mo:Zr mass ratio. However, the measured excesses did not match the expected fission-induced pattern, especially for ^{98}Mo. It is conceivable that this discrepancy was due to Ru interferences from the ^{96,98,100}Ru isotopes. This constitutes a source of uncertainty which was not accounted for and could have impacted on the measured ^{96}Mo excess, leading to a shorter half-life. In this work particular care was exercised to take into account Ru interferences. In addition, the Mo:Zr mass ratio was directly measured to confirm that the measured excesses were genuinely related to the decays.

III. MEASUREMENT METHODS

A. Reagents, samples, and equipment

A zircon reference material (Geological Survey of Canada reference, herein labeled as GSC-1242) was obtained from the Lac Fréchette pluton in Western Quebec, Canada. Standard mineral separation techniques including crushing, water-table separation, heavy-liquids, and Frantz$^{\rm TM}$ magnetic separation were used to obtain a zircon concentrate from the syenite [14]. Zircon grains were pale to dark pinkish-brown, clear, and showed stubby prismatic to equant multifaceted morphology. Internal fracturing resulting from volume expansion during metamictisation of the zircon crystal lattice was evident throughout the zircon sample. Dating of the syenite by ID-TIMS demonstrated the isotopic homogeneity of the material and yielded an age of $2681.0^{+1.9}_{-1.4}$ Myr [15]. Uranium concentrations were measured by laser-ablation ICPMS following the methods in Ref. [14]. A sample of 38 zircons were measured against a NIST SRM 610 glass along with an internal zircon reference material. The U concentrations varied from 75–200 μg/g, with a mean value of 172(15) μg/g.

In addition, younger detrital zircon samples from the Yoganup Strand Line were obtained from a Westralian Sands Limited (WSL) mineral sand mining operations at Capel, Western Australia. Within the sample population, individual zircon ages ranged from 150 to nearly 3500 Myr, and a mean age of 910(30) Myr was obtained for the bulk sample based on 510 ages.

Solutions were prepared with high-purity reagents including Seastar$^{\rm TM}$ Baseline® 47–51% hydrofluoric acid (HF), Anachemia Environmental Grade Plus 32–35% hydrochloric acid (HCl), and BDH Aristar® Ultra 67–70% nitric acid (HNO_3). Reagents were diluted with Milli-Q® water purified to 18 M$\Omega \cdot$cm. Measurements were calibrated to dilutions of ICP standards: Specpure® 1000 μg/g Zr and PlasmaCal® 10000 μg/g Mo. All dilutions and other mass measurements were performed with a calibrated Mettler-Toledo AT201 analytical balance (precision 10 μg).

Acid digestions were performed in a custom HF-resistant Parr Instrument Company model-4746 high-pressure

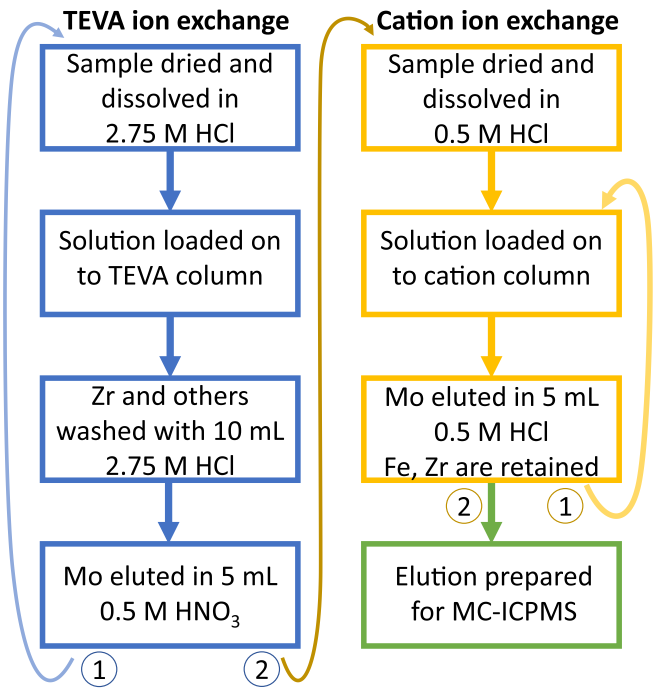

FIG. 2. Flow chart summarizing ion-exchange separation of Mo from Zr, Fe, and other elements. Removal of Fe is important as it results in an interference at $A = 96$ from $^{56}\mathrm{Fe}^{40}\mathrm{Ar}^{+}$. Two successive separations using TEVA resin remove Zr by four to six orders of magnitude, followed by two successive separations using cation resin to remove Fe and Zr by an additional three to four orders of magnitude.

acid-digestion vessel constructed from the nickel-copper alloy 400 (Monel®). Ion exchanges were performed with Eichrom® tetravalent (TEVA) resin (mesh size 50–100 μm) and Eichrom® analytical grade cation-exchange resin (50W × 8, 100–200 mesh). Mass-spectrometry measurements were carried out on a Thermo Scientific Neptune™ multicollector inductively coupled plasma mass spectrometer (MC-ICPMS) equipped with nine Faraday cups, a secondary-electron multiplier, and multi-ion counting (MIC) detectors. Samples were introduced through an Elemental Scientific Apex-Q desolvating nebulizer with a 130 μL/min PFA nebulizer.

B. Chemical separation of Mo from $ZrSiO_4$

To measure the isotopic composition of the Mo contained in the zircon sample, it must first be separated from the Zr. This is challenging as there is a factor of $> 10^6$ more Zr than Mo in the sample. A complete description of the acid digestion and ion-exchange development can be found in Ref. [16]. To overview, bulk zircon samples were dissolved by HF acid digestion in high-pressure acid-digestion vessels. A 492.6-mg aliquot of the GSC-1242 zircon sample was digested in 12.83 g of concentrated HF at 215 °C for 96 hours. Zr recovery of 63(3)% was achieved, yielding a solution with 156(8) mg Zr as measured by ICPMS.

Mo was then isolated by ion-exchange chemistry with four successive ion exchanges as shown in Fig. 2. The final Zr content was measured by ICPMS to be 1.4 ng, which is an eight orders of magnitude reduction, while the Mo content was measured to be 27(5) ng after separation from $ZrSiO_4$. The zircon sample Mo:Zr mass ratio was therefore evaluated to $m_{\mathrm{Mo}}/m_{\mathrm{Zr}} = 173(32)$ ng/g.

A full replicate digestion and separation was performed on a second GSC-1242 sample. The digested solution contained 129(7) mg Zr and 23(4) ng Mo, yielding a Mo:Zr mass ratio of 181(33) ng/g.

C. Mass spectrometry of Mo

Purified Mo from the zircon sample was dissolved in 0.5 mL of 0.5 M HNO_3 and was measured along with dilute ICP-standards of Mo and Zr with a Thermo Scientific Neptune MC-ICPMS. The $^{92-98}$Mo isotopes were measured on Faraday cups L2-H3 equipped with 10^{11} Ω amplifiers, while ^{90,91}Zr were monitored on Faraday cups L4 and L3 equipped with 10^{12} Ω amplifiers and ^{99}Ru was monitored on an ion counter attached to H4. Each sample was measured over 60 cycles of 2-second integrations, and a set of corrections were performed offline [16]. The measurement session included measurements of 10–200 ppb Mo ICP standards, a 50 ppb Zr ICP standard to improve the Zr correction, and a mixture of 5 ppb Zr and 50 ppb Mo ICP standards. This last constituted a similar composition as the purified Mo sample and was included to verify the effectiveness of Zr correction. Prior to each sample measurement, an on-peak blank measurement was performed to allow for "blank" subtraction of background contaminants such as $^{40}\mathrm{Ar}_2{}^{16}\mathrm{O}^{+}$.

A data analysis algorithm was developed in MATHEMATICA® which is described in detail in Ref. [16]. It consists of a set of data filters, a "blank" subtraction to remove spectral interferences, and corrections for Zr and Ru interferences. Finally, each measurement was corrected for mass fractionation based on the measured n(^{97}Mo)/n(^{95}Mo) ratio of the 200 ppb Mo ICP standard. This ratio was chosen as the isotopes are free from interferences from Zr and Ru. Uncertainties were estimated based on the fluctuations of the Mo isotope ratios in the corrected data and the uncertainties of the interference corrections. These uncertainties were added in quadrature.

As shown in Table I, all isotopic-composition measurements of the ICP standards and mixtures showed no significant deviation from the initial ICP standard. Further, the measurement of Mo purified from a younger 0.9 Gyr zircon sample showed no deviation. This sample was expected to not have any resolvable excesses due to its younger age and 60% higher Mo content of 272(40) ng/g. With typical uncertainties of less than 50 ppm (1σ), this demonstrated the effectiveness of the present analytical method. The δ^{98}Mo value of the 2.68 Gyr zircon sample showed a clear deviation from the ICP standard. This was due to the spontaneous fission (SF) of natural uranium contained in the zircon sample decaying to ^{95,97,98}Mo, affecting the fractionation correction.

D. Mass-dependent fractionation correction adjusted for spontaneous fission of uranium

The mass-dependent fractionation of Mo from the 2.68 Gyr zircon sample cannot be accurately corrected based on the

TABLE I. Measured isotopic compositions of laboratory ICP standards, the 0.9 Gyr zircon sample, and two full replicate 2.68 Gyr zircon samples. Column one: Samples measured, where the numbers in square brackets denote the element concentration in ng/g; column two to four: measured concentrations of Zr, Ru, and Mo based on ion current intensity; column five to seven: δ values of the n(^{X}Mo)/n(^{95}Mo) ratios relative to the initial Mo ICP standard. Isotopic compositions are corrected for mass-dependent fractionations by normalizing to the n(^{97}Mo)/n(^{95}Mo) ratio. *Zircon δ values calculated relative to the SF-adjusted (SFA) Mo laboratory standard.

Sample ID	[Zr] (ng/g)	[Ru] (pg/g)	[Mo] (ng/g)	δ^{92}Mo (ppm)	δ^{94}Mo (ppm)	δ^{96}Mo (ppm)	δ^{98}Mo (ppm)
Mo[200]	0.01	0.33	198	1(12)	10(9)	10(8)	13(12)
Mo[50]	0.43	<0.1	54	−10(30)	5(19)	10(14)	8(23)
Mo[10]	0.01	<0.1	11	10(70)	−60(60)	34(29)	−30(40)
Mo[50] Zr[5]	49	<0.1	55	−52(30)	20(40)	37(9)	22(18)
Zircon (0.9 Gyr)	0.15	2.8	21	−40(50)	−2(50)	−8(21)	7(23)
Zircon-A (2.68 Gyr, no SFA)	1.1	17	16	20(70)	−40(60)	−11(27)	−110(40)
Zircon-A (2.68 Gyr, SFA)*	**1.1**	**17**	**16**	**45(70)**	**31(60)**	**107(40)**	**4(50)**
Zircon-B (2.68 Gyr, SFA)*	**0.004**	**10**	**38**	**−9(20)**	**9(20)**	**88(20)**	**7(25)**

$n(^{97}\text{Mo})/n(^{95}\text{Mo})$ of the laboratory ICP standard. This is due to the contribution from spontaneous fission (SF) of U leading to ^{95,97,98}Mo over the geological time scales. Further, the n(^{94}Mo)/n(^{92}Mo) ratio cannot be used either because of its larger uncertainty as a result of the Zr interference. The amount of U SF products can, however, be evaluated using the cumulative spontaneous fission yields of ^{95,97,98}Mo from the database of Ref. [17]: 6.11%, 6.62%, and 6.40%, respectively, with each yield having a relative uncertainty of 2% [18]. The U content of the zircon sample was measured to be 172(15) μg/g of ZrSiO$_4$, and the Mo content was 173(32) ng/g of Zr. One can therefore adjust the reference isotopic composition of Mo to account for the U SF contribution.

Table I and Fig. 3 show the measured δ-values of Mo from the 2.68 Gyr zircon sample, where the adjusted laboratory standard was used as the reference. The $n(^{92,94,98}\text{Mo})/n(^{95}\text{Mo})$ ratios were all within uncertainty of the SF-adjusted standard, demonstrating the effectiveness of the corrections, while the $n(^{96}\text{Mo})/n(^{95}\text{Mo})$ ratio showed a clear excess at a level of 107(40) ppm. A full replicate analysis, Zircon-B, yielded an excess of 88(20) ppm.

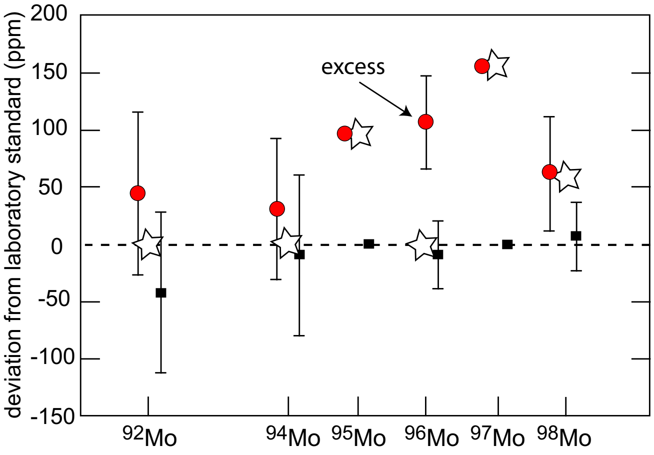

FIG. 3. Shown in red circles are the isotopic excesses of the Mo element in the 2.68 Gyr old zircon sample. Stars indicate predicted excesses as a result of spontaneous fission of trace amounts of natural U in the zircon sample, while squares show the isotopic excesses measured in a 0.9 Gyr zircon sample. The $n(^{97}\text{Mo})/n(^{95}\text{Mo})$ ratio was fixed to correct mass-dependent fractionation.

IV. ^{96}Zr HALF-LIFE

As discussed in Ref. [12], there should be no sources of excess ^{96}Mo other than from the decay of ^{96}Zr. We surveyed further possible reactions, which could affect the isotopic composition of molybdenum within the ancient zircon sample. The nucleus ^{96}Zr does not feature a long-lived isomer, which could possibly be excited by some ambient nuclear reaction. Therefore, a competing β-decay to ^{96}Nb from such an isomer is excluded. A conceivable $^{96}\text{Zr}(p,n)$ reaction leading directly to ^{96}Nb requires proton energies well above 10 MeV to overcome the Coulomb barrier. Although muons from cosmic ray interactions are a potential source of protons of this energy when they are hitting the Earth's surface, their intensities are too low to produce an effect [19]. Further, a neutron induced β-decay of the form $n + (n) \rightarrow n + (p^+) + e^- + \bar{\nu}_e$, where the brackets indicate the bound nucleon, has far too small a cross section to be of any significance at all.

By applying the measured excesses along with $m_{\text{Mo}}/m_{\text{Zr}} =$ 173(32) ng/g and 181(32) ng/g, respectively, to Eq. (1), one calculates n_d/n_0 to be $1.05(30)\times10^{-10}$ and $0.89(18)\times10^{-10}$, respectively. Taking the weighted average value of $0.92(17)\times10^{-10}$ and the zircon age of 2.681(2) Gyr, one arrives at a half-life for ^{96}Zr according to Eq. (2) of

$$T_{1/2} = \left(2.03^{+0.46}_{-0.31}\right) \times 10^{19}\text{yr}. \tag{3}$$

An uncertainty budget analysis indicates that 50% of the uncertainty is due to δ^{96}Mo, 35% is due to the U concentration, while the remaining parameters contribute 0–8% each. The precision obtained for the δ value is close to the limit of what is achievable by MC-ICPMS, as the manufacturer guaranteed precision of 5–10 ppm is only possible with higher concentration, longer measuring times, and pure reference materials. An improvement in precision can therefore only be obtained with a larger sample of ancient, i.e., >2 Gyr, zircon with a significantly lower natural Mo content to increase the relative size of the decay excess.

It should be noted that a lower limit for the ^{96}Zr half-life can also be determined from the 0.9 Gyr zircon measurement. By likewise applying corrections for the spontaneous fission of ^{238}U, the corrected excess for the 0.9 Gyr zircon is $\delta^{96}\mathrm{Mo} = 9(21)$ ppm. This corresponds to a lower limit of $T_{1/2} \geqslant 1.4 \times 10^{19}$ yr, which is in agreement with the 2.68 Gyr zircon measurement.

The total decay rate determined by this measurement is due to both the single and double-β decays of ^{96}Zr:

$$\lambda_{\mathrm{net}} = \lambda_{\beta} + \lambda_{\beta\beta} \tag{4}$$

with the total decay rate $\lambda_{\mathrm{net}} = (3.42 \pm 0.63) \times 10^{-20}$ yr from this experiment, and $\lambda_{\beta\beta} = (2.95 \pm 0.14) \times 10^{-20}$ yr from that found in Ref. [13]. The single β decay rate is therefore $\lambda_{\beta} = (0.47 \pm 0.64) \times 10^{-20}$ yr, which is within uncertainty of zero. Therefore a 1σ lower limit on the single β decay half-life can be set as

$$T_{1/2}^{\beta} \geqslant 6.2 \times 10^{19}\mathrm{yr}. \tag{5}$$

This limit is a factor of 2 lower than the expected value from theoretical shell-model calculations [$T_{1/2}^{\beta} = (11 g_{\mathrm{A}}^{-2}) \times 10^{19}$ yr] and QRPA-model calculations [$T_{1/2}^{\beta} = (24 g_{\mathrm{A}}^{-2}) \times 10^{19}$ yr] quoted in Refs. [4,7], and it still lacks the precision to differentiate between these two models. As already indicated, the exact value of the axial-vector coupling constant g_A in the theoretical treatment of the weak interaction in many-body systems is unknown, especially for the higher-order forbidden decays, and it is either around 0.9 (quenched) or at the free value of 1.27.

V. OUTLOOK: QUADRUPLE β-DECAY OF ^{96}Zr → ^{96}Ru

The geochemical analytical methods described in this article can also be applied to a measurement of a ^{96}Zr to ^{96}Ru quadruple-β (4β) decay. This decay is of interest as the neutrinoless decay mode is possible also with Dirac neutrinos [20], and because of phase-space considerations it is even the most favorable decay mode. A few cases have been identified in the nuclear chart, where this decay is energetically possible. Of these, the isotope ^{96}Zr is the only system, where all intermediate nuclei have a lower mass, a fact which could potentially accelerate the decay. The ^{96}Zr 4β-decay Q value is, however, only 641.5 keV, which is relatively low compared to typical $\beta\beta$-decay Q values. The nucleus with the highest $Q_{4\beta}$ value of 2084 keV is ^{150}Nd, and for this the NEMO collaboration has published a lower bound of its 4β-decay half-life of $>1 \times 10^{21}$ yr [21].

In the present analysis the total ruthenium content in the GSC-1242 zircon sample was approximately 950 times less than the Mo content, which makes the Ru isotopic composition significantly more sensitive to radiogenic excesses. A sufficiently large quantity of >2 Gyr zircon material with a Ru content of <0.5 ppb could yield a lower limit for the 4β-decay half-life of order 10^{23} yr. This is possible because the isotope ^{96}Ru has several advantageous properties, as it features a low natural isotopic abundance of 5.5%, it is not produced in a U fission process, and it is also not produced via neutron capture from ^{95}Ru, since ^{95}Ru is unstable with a half-life of 1.6 h.

VI. CONCLUSION

A geochemical measurement of the ^{96}Zr half-life is performed, resulting in a value of $T_{1/2} = (2.03^{+0.46}_{-0.31}) \times 10^{19}$yr. This measurement applies modern isotope-geochemical instrumentation and chemical separation techniques to significantly improve the reliability of the results, taking into account all known interferences and sources of uncertainty. In particular, the effect of contamination due to ^{96}Ru is measured and corrected, as it likely contributes to the shorter ^{96}Zr half-life result from Ref. [12]. The overall ^{96}Zr-decay half-life is compared to the previously measured $\beta\beta$-decay half-life to set a lower limit for the single β-decay half-life of $T_{1/2}^{\beta} \geqslant 6.2 \times 10^{19}$yr. The limit is still too low to distinguish between models for theoretical calculations, but resolves the speculation of a much shorter half-life based on previous isotope abundance measurements.

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