Single and Double Beta-Decay Q Values among the Triplet ^{96}Zr, ^{96}Nb, and ^{96}Mo

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The atomic mass relations among the mass triplet ^{96}Zr, ^{96}Nb, and ^{96}Mo have been determined by means of high-precision mass measurements using the JYFLTRAP mass spectrometer at the IGISOL facility of the University of Jyväskylä. We report Q values for the ^{96}Zr single and double β decays to ^{96}Nb and ^{96}Mo, as well as the Q value for the ^{96}Nb single β decay to ^{96}Mo, which are $Q_\beta(^{96}\mathrm{Zr}) = 163.96(13)$, $Q_{\beta\beta}(^{96}\mathrm{Zr}) = 3356.097(86)$, and $Q_\beta(^{96}\mathrm{Nb}) = 3192.05(16)$ keV. Of special importance is the ^{96}Zr single β-decay Q value, which has never been determined directly. The single β decay, whose main branch is fourfold unique forbidden, is an alternative decay path to the ^{96}Zr $\beta\beta$ decay, and its observation can provide one of the most direct tests of the neutrinoless $\beta\beta$-decay nuclear-matrix-element calculations, as these can be simultaneously performed for both decay paths with no further assumptions. The theoretical single β-decay rate has been re-evaluated using a shell-model approach, which indicates a ^{96}Zr single β-decay lifetime within reach of an experimental verification. The uniqueness of the decay also makes such an experiment interesting for an investigation into the origin of the quenching of the axial-vector coupling constant g_A.

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The neutrinoless double beta ($0\nu\beta\beta$) decay is currently of significant interest in nuclear and particle physics. An observation of this decay mode not only gives insight into the nature of the neutrino but also provides information about its absolute mass scale. However, the theoretical description of the $0\nu\beta\beta$ decay appears to be highly nontrivial. The critical quantity which enters into these model calculations is the nuclear matrix element (cf. Refs. [1–3]). It describes the underlying nuclear physics and, because of its complexity, neither the matrix elements nor the adequacy of the models can be easily assessed. In this context, ^{96}Zr is of special interest, as it offers a rather unique opportunity to experimentally test the predictive power of these models.

In $\beta\beta$ decay, ^{96}Zr features the third largest Q value, topped only by ^{48}Ca ($Q_{\beta\beta} = 4.268$ MeV) and ^{150}Nd ($Q_{\beta\beta} = 3.371$ MeV) [4,5]. In addition, ^{96}Zr is also unstable against single β decay, which is a property it only shares with ^{48}Ca. If this decay is observed in either of these systems, a comparison of the respective measured and theoretical single β-decay rate allows a direct test of the nuclear-matrix-element calculations for $\beta\beta$ decay, as these follow the same theoretical prescription. However, as far as ^{48}Ca is concerned, shell-model calculations [6,7] suggest that its single β-decay half-life is of the order 10^{21} yr, which is more than an order of magnitude longer than the one for the $\beta\beta$ decay. The low isotopic abundance makes ^{48}Ca an even more unfavorable test case, which leaves ^{96}Zr, because of a more advantageous phase-space factor, as the only practical case where a measurement of the single β-decay half-life could be attempted.

Figure 1 shows the possible decay scheme of ^{96}Zr for single and double β decay. Single β decay is energetically possible into the $J^\pi = 6^+$ ground state, the $J^\pi = 5^+$, 44.2 keV first excited state, and the $J^\pi = 4^+$, 146.1 keV second excited state of ^{96}Nb. Among these, the decay to the

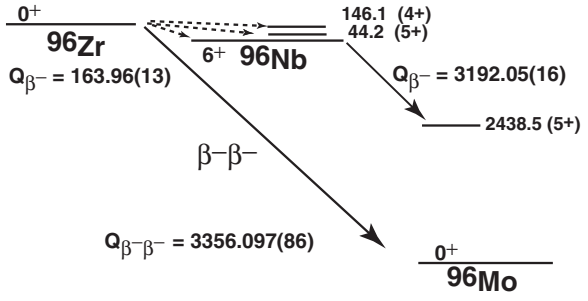

FIG. 1. Decay scheme for the $A = 96$ triplet showing the energy position of ^{96}Zr with respect to its neighbors ^{96}Nb and ^{96}Mo. The Q values are from this work (all energies are in keV).

$J^\pi = 5^+$, 44.2 keV excited state is fourfold unique forbidden and considered the one with the highest decay probability exceeding the other two, which are sixfold and fourfold nonunique forbidden, by several orders of magnitude [8].

Of all possible decays only the partial half-life for the 2ν variant of the $\beta\beta$ decay to the ^{96}Mo ground state is sufficiently known from the NEMO-3 Collaboration, which is $T^{\beta\beta}_{1/2} = (2.3 \pm 0.2) \times 10^{19}$ yr [9,10]. An earlier geochemical measurement of the ^{96}Zr decay rate was reported in Ref. [11], where the total half-life was determined from the isotopic abundance anomaly of ^{96}Mo in ancient zircon samples originating from Australia. Note that ^{96}Mo is the final stable nucleus in both decay chains. This measurement yielded a rather short value of $T_{1/2} = (0.94 \pm 0.32) \times 10^{19}$ yr. Although systematic effects are difficult to control in geochemical measurements, the low value can be reconciled with the NEMO-3 result if the ^{96}Zr $\beta\beta$ decay is appreciably contaminated by two sequential single β decays as indicated in Fig. 1. In that case the single β-decay half-life may be evaluated to $T^{\beta}_{1/2} = (1.6 \pm 0.9) \times 10^{19}$ yr. An even earlier geochemical measurement reported in Ref. [12] gave a total half-life of $T_{1/2} = (3.9 \pm 0.9) \times 10^{19}$ yr. This value is not compatible with the NEMO-3 results.

Direct single β-decay half-life measurements for ^{96}Zr have so far only established lower bounds. The strongest one is $T^{\beta}_{1/2} > 2.6 \times 10^{19}$ yr [13], which supersedes an earlier value given in Ref. [14] as a result of a more accurate accounting of the γ-ray multiplicity when evaluating the efficiency of the setup. A more recent and slightly less competitive limit is published in Ref. [15] at $T^{\beta}_{1/2} > 2.4 \times 10^{19}$ yr.

A theoretical value for the half-life is given at $T^{\beta}_{1/2} = 24 \times 10^{19}$ yr in Ref. [8], where the fourfold ($L = 4$) forbidden matrix element was calculated in the quasiparticle random phase approximation (QRPA), and the phase-space factor assumed a ground-state to ground-state Q value of 163 keV. On the other hand, the QRPA also appears as the model employed for describing the $0\nu\beta\beta$-decay matrix elements. In this case the high-momentum transfer (≈ 0.5 fm^{-1}) at the neutrino vertices relaxes the degree of forbiddenness and allows for multipoles of up to $L \approx 6$ in the transition amplitudes [16]. A test of the QRPA —and consequently a test of its applicability to $\beta\beta$ decay— could be performed if the two single β-decay quantities, i.e., the decay rate and the Q value, were known, because these determine the experimental ft value and the single β-decay matrix element. One may note that the phase-space factor for a fourfold forbidden decay depends in leading order on Q^{13}, which makes the transition particularly sensitive to the Q value. Furthermore, the uniqueness of the $^{96}\mathrm{Zr}(J^\pi = 0^+) \rightarrow {}^{96}\mathrm{Nb}(44\ \mathrm{keV}, J^\pi = 5^+)$ transition ensures that there is only one matrix element active, thereby providing a particularly clean test of the underlying model.

The only ^{96}Zr single β-decay Q-value determination goes back to a ^{96}Nb β-decay end-point energy measurement [17], where the decay leads to the $J^\pi = 5^+$ level at 2438.5 keV in ^{96}Mo (cf. Fig. 1). This measurement has so far also been the sole reference for the evaluation of the absolute mass of ^{96}Nb in the 2012 Atomic Mass Evaluation (AME12) [5]. The measured β spectrum contained, however, two extra transitions which were included in the final analysis. The origin of one still remains unknown, whereas the other later turned out to be inconsistent with the ^{96}Nb level scheme. Furthermore, a recently performed high-precision Penning trap measurement of the ^{96}Zr $\beta\beta$-decay Q value [18] gave a nearly 7 keV higher value than the one in the AME12 [5] (i.e., a 3σ deviation). This 7 keV shift not only is of critical importance for the $0\nu\beta\beta$-decay signal extraction in present $\beta\beta$-decay experiments, but it may also have an impact on the single β-decay Q value. A re-examination of the mass relations among the $A = 96$ triplet—and in particular a precision determination of the single β-decay Q value of ^{96}Zr—was therefore called for.

The measurements were performed at the IGISOL facility [19,20] of the University of Jyväskylä. A 57% enriched ^{96}Zr metal foil with an areal thickness of 1 mg/cm^2 was bombarded with an ≈ 2 μA proton beam at 10 MeV. The ^{96}Nb isotope was produced by a (p,n) reaction and both isobars, ^{96}Nb and ^{96}Zr, were released from the target with comparable intensities. In addition, the Havar® primary beam window and also the target stainless steel mounting frame constitute a molybdenum containing alloy, from which also the ^{96}Mo isobar was released with similar intensities. This fortuitous situation allowed a simultaneous measurement for all three isobars, thereby minimizing the potential systematic effects. Prior to the on-line measurements, mass measurements were performed with the stable isotopes ^{96}Zr and ^{96}Mo from the off-line discharge ion source, which provided an intensity ratio of 1:10.

The ions were thermalized in the IGISOL gas cell and were transported by means of gas flow and the sextupole ion guide to the high-vacuum region, where they were accelerated with a 30 kV potential and mass number selected with a dipole magnet. The $A/q = 96$ ions were injected into the radio frequency quadrupole cooler and buncher [21] and then transferred to the JYFLTRAP system [22]. The JYFLTRAP features two cylindrical Penning traps in a 7 T magnetic field. The first trap is the purification trap filled with helium buffer gas at low pressure. This trap is used for producing an isobarically purified beam by means of the buffer-gas cooling technique [23]. The second trap is the precision mass-measuring trap operating under high vacuum ($p < 10^{-7}$ mbar), where the cyclotron frequency of the injected ion is determined by the time-of-flight ion-cyclotron-resonance technique (TOF-ICR) [24].

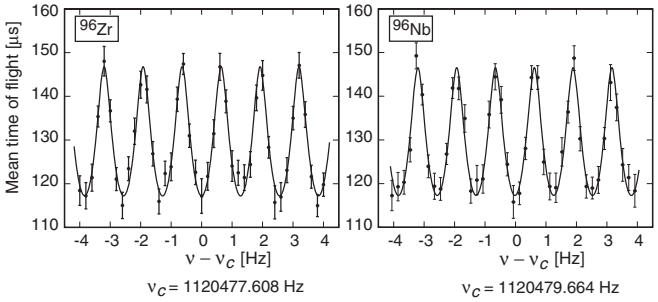

FIG. 2. Time-of-flight spectrum for the (^{96}Zr,^{96}Nb) pair using a 25-750-25 ms (on-off-on) Ramsey excitation pattern. The solid line represents a fit to the data using the theoretical line shape as described in Ref. [26].

The mass doublet ^{96}Zr and ^{96}Nb exhibits a mass difference of ≈164 keV, which translates into a cyclotron-frequency difference of ≈2 Hz. This difference is too small for employing the buffer-gas cooling technique and necessitates a more elaborate Ramsey cleaning, which is described in detail in Ref. [25]. For the actual mass measurement, a Ramsey excitation pattern of 25-750-25 ms (on-off-on) was employed for the TOF-ICR measurement (see Fig. 2). The Ramsey excitation method is described in, for instance, Refs. [27,28].

The measurements were performed by switching between the ion species in the pairs (^{96}Zr,^{96}Nb), (^{96}Nb,^{96}Mo), and (^{96}Zr,^{96}Mo) (the latter off-line) after each scanning cycle, whereby a scanning cycle took about a minute to complete. In the analysis, typically 10–20 rounds were summed before a time-of-flight fit was performed and the cyclotron frequencies $\nu_c^{(i)}$ of the pairs and their ratios R,

$$R = \nu_c^{(1)}/\nu_c^{(2)}, \qquad \nu_c^{(i)} = \frac{1}{2\pi}\frac{eB}{m_i}, \tag{1}$$

were evaluated. By this mode of operation, fluctuations of the magnetic field, which are below $\Delta B/B = 10^{-11}$ per minute, need not be considered, and since each of the ion pairs constitutes an A/q doublet, systematic effects resulting from field imperfections cancel in the frequency ratio [29]. Furthermore, no systematic effects due to ion-ion interactions were seen when the data were analyzed using a count-class analysis as described in Ref. [30].

The Q value for each pair is determined as

$$Q_{21} = M_2 - M_1 = (R-1)(M_1 - m_e), \tag{2}$$

where m_e is the electron mass and M_2, M_1 are atomic masses of the β-decay parent and daughter, respectively. The electron binding-energy difference is about a tenth of an eV and is neglected in Eq. (2).

Figure 3 shows the sequences of the Q-value measurements as a function of the elapsed time for each pair of the $A = 96$ triplet together with the distribution of the individual measurements. The final results are given in Table I.

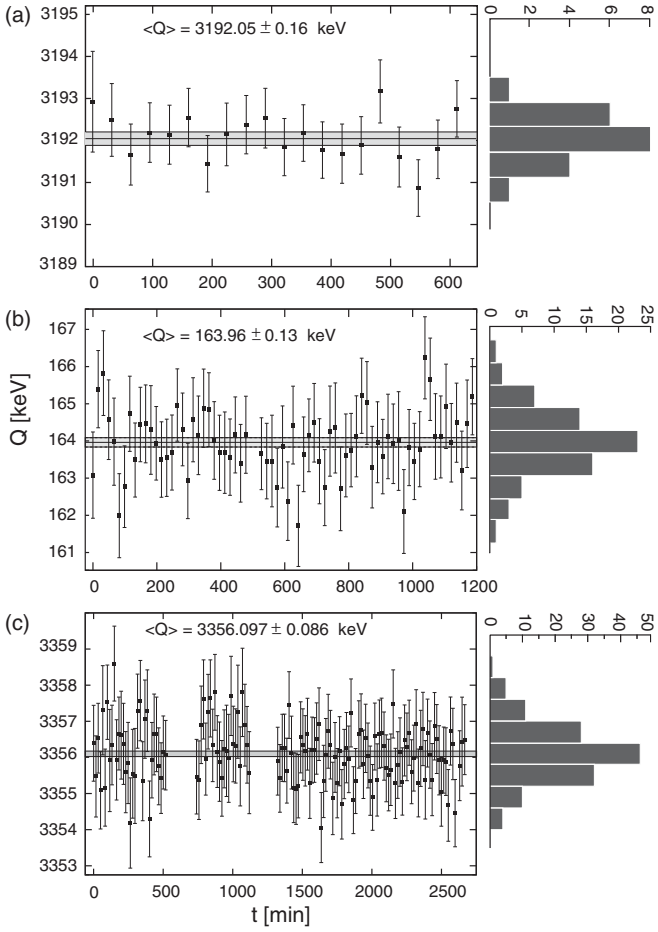

FIG. 3. Q-value results for (a) ^{96}Nb to ^{96}Mo, (b) ^{96}Zr to ^{96}Nb, and (c) ^{96}Zr to ^{96}Mo decays. The individual measurements are plotted against the elapsed time of the experiment. The grey bands indicate the final errors around the central values. Also shown is the distribution of the individual measurements within ±300 eV bins indicating a normal distribution.

We note that, as an independent check, the triangle relation for the central values

$$Q_{\beta\beta}(^{96}\mathrm{Zr}) = Q_\beta(^{96}\mathrm{Zr}) + Q_\beta(^{96}\mathrm{Nb}) \tag{3}$$

is satisfied to within 87 eV (left-hand-side error 86 eV, right-hand-side error 206 eV). Furthermore, Table I contains the Birge ratios [31] for each measurement, which indicates that the statistical errors for extracting the various Q values are even slightly overrated by ≈23%.

The $\beta\beta$-decay Q value of the present measurement is higher than the one given in Ref. [18] by 0.25 keV, though still marginally consistent at about a 1σ level. Compared to the Q value listed in the AME12 [5], it has increased by 7.1 keV and is now evaluated at $Q_{\beta\beta} = 3356.097$ keV, with an accuracy of 86 eV. The Q value for the ^{96}Zr single β decay comes to 163.96 keV, with an accuracy of 130 eV. Furthermore, taking the mass-excess value of ^{96}Mo from the AME12 as a reference (i.e., −88793.6(04) keV), the measured Q values fix the absolute mass values of ^{96}Nb

TABLE I. Measured cyclotron-frequency ratios (here, $R-1$) for the $A=96$ mass triplet, Q values, Q-value uncertainties ΔQ, and the Birge ratios for each measurement. Each Q value is the weighted mean of the individual measurements appearing in Fig. 3. The lower part of the table lists the mass excess (ME) for each isobar, where the ^{96}Mo value is taken as a reference value from Ref. [5], and its uncertainty is given in the second set of parentheses. The differences between the present mass-excess values and those from the AME12 [5] are listed as well.

Isobaric pair (M_1/M_2)	$R-1$ (10^{-9})	Q (keV)	ΔQ (keV)	Birge ratio
^{96}Mo/^{96}Zr	37567.92(096)	3356.097	0.086	0.79
^{96}Nb/^{96}Zr	1835.34(140)	163.96	0.13	0.78
^{96}Mo/^{96}Nb	35731.53(179)	3192.05	0.16	0.75

Nucleus	ME (keV)	ME (Ref. [5]) (keV)	Diff. (keV)
^{96}Mo		−88793.6(04)	
^{96}Nb	−85601.46(16)(40)	−85607.0(30)	$+5.53\pm3$
^{96}Zr	−85437.50(09)(40)	−85444.6(20)	$+7.10\pm2$

and ^{96}Zr as well. These exhibit up to a 3.5σ deviation from the present AME12 values and about a 20 times higher accuracy (cf. the lower part of Table I).

Theoretical model calculations were performed for all possible ^{96}Zr single β transitions using the formalism outlined in Ref. [32]. The nuclear matrix elements were computed in the framework of the nuclear shell model employing the shell-model code NuShellX [33]. The Gloeckner interaction [34] was adopted in a model space consisting of single-particle orbits $2p_{1/2}$ and $1g_{9/2}$ for protons and $3s_{1/2}$ and $2d_{5/2}$ for neutrons. The computed level scheme for ^{96}Zr and ^{96}Nb turned out to be in remarkably good agreement with the one established by experiment. In particular, the model correctly predicts the order of the 6^+, 5^+, and 4^+ states in ^{96}Nb. Calculated excitation energies are 32 keV for the 5^+ state and 304 keV for the 4^+ state, to be compared with the experimental energies of 44 and 146 keV (cf. Fig. 1).

Previous nuclear shell-model studies [35] of β-decay rates in the pf shell suggest a quenched axial-vector coupling constant of $g_A\approx1$. Although the quenching issue is far from being settled [36–39], by following this suggestion and taking the experimental decay energies from the present Q value, the partial half-lives for the various $^{96}\mathrm{Zr}\rightarrow{}^{96}\mathrm{Nb}$ β transitions are calculated as (i) $T^{\beta}_{1/2}(6^+)=1.6\times10^{29}$ yr, (ii) $T^{\beta}_{1/2}(5^+)=11\times10^{19}$ yr, and (iii) $T^{\beta}_{1/2}(4^+)=7.5\times10^{22}$ yr. Clearly, the total single β-decay half-life is dictated by the fourfold forbidden transition to the 5^+ state, thus confirming the conclusion of Ref. [8]. Furthermore, because only one nuclear matrix element is involved in this transition, one can cast the single β-decay half-life in the form $T^{\beta}_{1/2}=(11g_A^{-2})\times10^{19}$ yr. This value is about a factor of 2.2 smaller than the one from previous QRPA calculations [8], and the difference is almost entirely related to the difference in the nuclear-structure models. In fact, a re-evaluation of the QRPA calculations of Ref. [8] showed that the present Q value can only account for a 6.5% change. This renders an experimental half-life determination even more imperative.

Of course, it would be desirable to compute the $2\nu\beta\beta$-decay rate of ^{96}Zr in the same framework and compare with experiment to further assess the reliability and adequacy of the present single β-decay calculations. Unfortunately, in the Gloeckner single-particle space, this is not possible since no 1^+ intermediate states in ^{96}Nb can be formed. Extensions of the model space with appropriate interactions are not trivial, which is why there are no reported shell-model results for the $2\nu\beta\beta$ decay of ^{96}Zr yet.

In conclusion, the mass differences between each pair of the isobaric triplet ^{96}Zr, ^{96}Nb, and ^{96}Mo have been measured at the IGISOL-JYFLTRAP facility to an accuracy on the order of 100 eV. The single β-decay Q value, which so far had remained in doubt, has now been established at 163.96(13) keV. As a side product, the $\beta\beta$-decay Q value was determined to 3356.097(86) keV, which is 7.1 keV higher than the one quoted in the AME12 and 0.25 keV higher than the most recent measurement. It has been argued that a measurement of the single β-decay rate of ^{96}Zr can provide one of the most direct tests of models aimed at predicting the nuclear matrix elements for its $\beta\beta$ decay. A shell-model calculation and a recent QRPA calculation are found to deviate in their prediction for the single β-decay rate by more than a factor of 2. We also argued that an experimental determination of the fourfold forbidden β-decay half-life complements the known $2\nu\beta\beta$-decay half-life. Since the former is proportional to g_A^{-2} and the latter to g_A^{-4}, a simultaneous theoretical evaluation of the participating nuclear matrix elements could shed light on the quenching of the axial-vector coupling constant.

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