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Determination of the direct double-β decay Q value of ^{96}Zr and atomic masses of $^{90-92,94,96}$Zr and $^{92,94-98,100}$Mo

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Experimental searches for the neutrinoless double-β decay offer one of the best opportunities to look for physics beyond the Standard Model. Detecting this decay would confirm the Majorana nature of the neutrino, and a measurement of its half-life can be used to determine the absolute neutrino mass scale. Important to both tasks is an accurate knowledge of the Q value of the double-β decay. The LEBIT Penning trap mass spectrometer was used for the first direct experimental determination of the ^{96}Zr double-β decay Q value: $Q_{\beta\beta}$ = 3355.85(15) keV. This value is nearly 7 keV larger than the 2012 atomic mass evaluation [Chin. Phys. C **36**, 1603 (2012)] value and one order of magnitude more precise. The 3-σ shift is primarily due to a more accurate measurement of the ^{96}Zr atomic mass: $m(^{96}\mathrm{Zr})$ = 95.90827735(17) u. Using the new Q value, the $2\nu\beta\beta$-decay matrix element, $|M_{2\nu}|$, is calculated. Improved determinations of the atomic masses of all other zirconium ($^{90-92,94,96}$Zr) and molybdenum ($^{92,94-98,100}$Mo) isotopes using both $^{12}C_8$ and ^{87}Rb as references are also reported.

I. INTRODUCTION

A confirmed observation of neutrinoless double-β ($0\nu\beta\beta$) decay offers many exciting opportunities to study new physics beyond the Standard Model. This decay requires a nonzero neutrino mass, which has been confirmed by neutrino oscillation experiments [1, 2]. Additionally required, but unconfirmed, is the Majorana nature of the neutrino, which would mean that the neutrino is its own antiparticle, and violation of lepton number conservation.

Current experimental searches for $0\nu\beta\beta$-decay have limited the half-life to the order $T_{1/2} \sim 10^{24} - 10^{25}$ y [3]. To account for such a rare process, these searches must incorporate large quantities of a source material to enhance the probability of detecting a decay event. In addition, background events must be supressed by sufficiently low contamination, or rejected by tracking techniques. Sources used for $0\nu\beta\beta$-decay searches must therefore have sufficient natural abundances, or enrichment techniques to provide the large amount of material of adequate purity. ^{96}Zr is one of the best candidates presently used in, or considered for, $0\nu\beta\beta$-decay searches [3], and the only one for which the Q value has not yet been measured by Penning trap mass spectrometry (PTMS).

A large decay Q value, given by the mass difference between the parent and daughter nuclei, is very important for the experiments. First, a large Q value separates the $0\nu\beta\beta$-decay peak from typical sources of background in the sum-energy spectrum of the emitted electrons. A large Q value also increases the decay probability by enhancing the $0\nu\beta\beta$-decay phase-space factor $G_{0\nu} \propto Q^5$. No experimental searches for the $0\nu\beta\beta$-decay are currently employing ^{96}Zr as a possible candidate, partly due to its relatively low abundance (2.8%). The ^{96}Zr $0\nu\beta\beta$-decay, however, has the second-largest Q value and third-largest phase-space factor of all double-β decay candidates [3]. In order to identify the $0\nu\beta\beta$-decay peak in the decay spectrum and confirm the event, the Q value must be accurately known to an uncertainty well within the energy resolution of the detector at that energy ($\sim$ 1%). For example, using a liquid scintillator containing a zirconium complex with an energy resolution of 4% at 2.5 MeV [4], a search of the 3.36 MeV signal from ^{96}Zr $0\nu\beta\beta$-decay would require a precision of $\sim$1 keV.

A precise Q value is also necessary for extracting the effective Majorana mass of the electron neutrino, $\langle m_{\beta\beta}\rangle$. If $0\nu\beta\beta$-decay is observed, and its half-life, $T^{0\nu}_{1/2}$, is obtained, then $\langle m_{\beta\beta}\rangle$ could be determined from the equation

$$(T^{0\nu}_{1/2})^{-1} = G_{0\nu}(Q_{\beta\beta}, Z)|M_{0\nu}|^2(\langle m_{\beta\beta}\rangle / m_e)^2, \qquad (1)$$

where Z is the nuclear charge, m_e is the electron rest mass, and $M_{0\nu}$ is the relevant nuclear matrix element.

PTMS has become a very important tool in mass measurements of atomic nuclei due to the high precision and accuracy that it provides [5–7]. In this paper, we present the first direct measurement of the ^{96}Zr double-β decay Q value using PTMS. The Q value is used to determine the $0\nu\beta\beta$-decay phase-space factor $G_{0\nu}$, the $2\nu\beta\beta$-decay phase-space factor $G_{2\nu}$ and the matrix element $|M_{2\nu}|$. Also presented in this paper are improved determinations of the atomic masses of all other stable Zr ($^{90-92,94,96}$Zr) and Mo ($^{92,94-98,100}$Mo) isotopes using both the $^{12}C_8$ carbon cluster and ^{87}Rb as references.

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II. EXPERIMENT DESCRIPTION

The portion of the Low-Energy Beam and Ion Trap (LEBIT) facility [8] used in this work is shown in Fig. 1. The main components of the LEBIT facility are a laser ablation ion source [9], a surface ionization source, a beam cooler and buncher based on a buffer gas-filled linear radiofrequency quadrupole (RFQ) ion trap [10], and a high-precision 9.4 T Penning trap mass spectrometer which utilizes the time-of-flight ion cyclotron resonance (TOF-ICR) detection technique [11].

Zr, Mo and $^{12}C_8$ ions are produced with a laser ablation ion source which employs a pulsed, frequency-doubled Nd:YAG laser. Targets, consisting of two semi-circular plates of natural zirconium/molybdenum, zirconium/carbon, or molybdenum/carbon were mounted on a rotatable holder inside a vacuum chamber. The targets were selectively rotated by a computer-controlled stepper motor such that only ions of the desired material were produced. Surface ionized $^{87}Rb^+$ is obtained by heating a tungsten filament of the plasma ion source that is located perpendicular to the LEBIT beamline and opposite to the laser ablation ion source.

After production, the ions were electrostatically accelerated to 5 keV and transported to the cooler/buncher through an electrostatic lens, which is also used to control the number of ions. Here ions were captured and cooled in a helium buffer gas at $\sim 2 \times 10^{-2}$ mbar then bunched at $\sim 10^{-3}$ mbar. The ions were then ejected as sub-microsecond pulses towards the 9.4 T Penning trap mass spectrometer.

The laser ion source for a given target material provided not only the particular ions of interest but also non-isobaric ions of the same material and other ions from target impurities. Therefore, purification was required prior to the mass measurements to avoid systematic errors due to ion-ion interactions in the Penning trap [12]. Non-isobaric contaminants were suppressed both by optimizing the radiofrequency amplitudes used in the cooler-buncher for the mass of interest and employing a fast electrostatic deflector in the beam line between the cooler/buncher and the Penning trap as a time-of-flight mass separator [13].

Once the ions were trapped, a strong dipolar radiofrequency (RF) pulse was applied at the reduced cyclotron frequencies of any possible contaminants to eject them from the trap as an added safeguard. The ions of interest were then excited by applying a quadrupolar RF electric field at a frequency near the cyclotron frequency of the ion [14]. A Ramsey excitation scheme [15, 16] was employed with two 150 ms long RF excitation pulses separated by 450 ms long wait time. The ions were then ejected from the trap and directed to a multi-channel plate (MCP) detector where the TOF was recorded. This MCP detector in Daly configuration has a measured efficiency of 63%. By varying the RF frequency, $\nu_{\rm RF}$, for multiple ion bunches of the same species, a TOF resonance curve was obtained, such as the $^{96}Zr^+$ resonance shown in Fig. 2. The theoretical line shape [17] has been fitted to the data to determine the frequency of the central minima, which occurs when $\nu_{\rm RF} = \nu_c$.

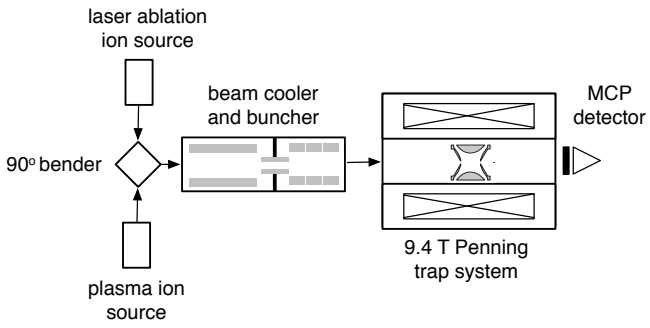

FIG. 1. Schematic of a subsection of the Low-Energy Beam and Ion Trap (LEBIT) Facility relevant to this article.

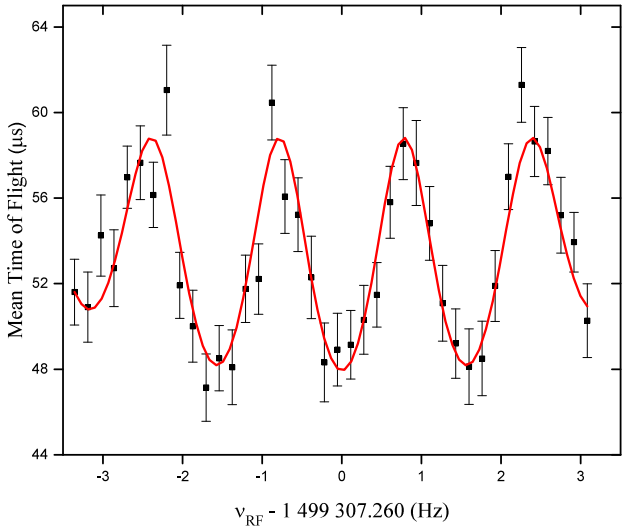

FIG. 2. (color online). One of 88 $^{96}Zr^+$ time-of-flight Ramsey cyclotron resonances used for the determination of $\nu_c(^{96}Zr^+)/\nu_c(^{96}Mo^+)$. This resonance contains about 1700 detected $^{96}Zr^+$ ions. The solid line is the theoretical line shape [17] fitted to the data.

III. Q VALUE DETERMINATION OF ^{96}Zr

The ^{96}Zr double-β decay Q value was determined from the cyclotron frequency ratio between the daughter and mother ions, $^{96}Mo^+$ and $^{96}Zr^+$. This value was obtained by performing alternating cyclotron frequency measurements of $^{96}Mo^+$ ions at time t_0, $\nu_c(^{96}Mo^+)(t_0)$, and time t_2, $\nu_c(^{96}Mo^+)(t_2)$, both before and after measuring the cyclotron frequency of $^{96}Zr^+$ at time t_1, $\nu_c(^{96}Zr^+)(t_1)$. To account for the magnetic field drifts, the ^{96}Mo cyclotron frequency measurements were linearly interpolated to determine the $^{96}Mo^+$ cyclotron frequency at time t_1, $\nu_c(^{96}Mo^+)(t_1)$ to give a frequency ratio of the two ions at the same time, $R = \nu_c(^{96}Zr^+)(t_1)/\nu_c(^{96}Mo^+)(t_1)$. That ratio is also equal to the inverse mass ratio,

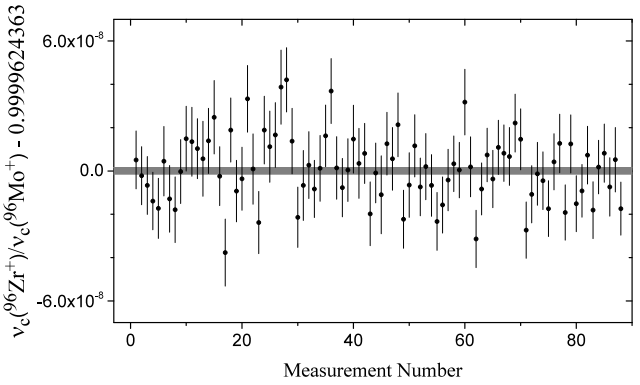

FIG. 3. Data sets of cyclotron frequency ratios $\nu_c(^{96}\mathrm{Zr}^+)/\nu_c(^{96}\mathrm{Mo}^+)$ about the set's mean value. The statistical uncertainty in an individual cyclotron frequency ratio is represented with the error bars, and the ± 1-σ statistical uncertainty of the data set is shown as a shaded band.

$m(^{96}\mathrm{Mo}^+)/m(^{96}\mathrm{Zr}^+)$, at (t_1).

Over a period of 110 hours, 88 cyclotron frequency ratio measurements were taken. Each ratio consisted of a TOF resonance of $^{96}\mathrm{Zr}^+$ bracketed by two resonances of $^{96}\mathrm{Mo}^+$. The data for each resonance took $\sim$ 35 minutes to gather and contained approximately 1700 detected ions. These cyclotron frequency ratios are shown in Fig. 3.

All known systematic frequency shifts amount to an insignificant error compared to the relative statistical uncertainty of 1.5×10^{-9}. Mass-dependent systematic effects in the cyclotron frequency ratio, such as those due to electric field imperfections or magnetic field inhomogeneities [18], are well below 1×10^{-10} [8] when comparing species with equal mass numbers. Nonlinear fluctuations in the magnetic field strength are minimized by stabilizing the pressure in the liquid helium cryostat of the solenoid magnet and lead to a relative error of 1×10^{-10} [19]. In addition, systematic frequency shifts can occur from Coulomb interactions between ions of the same species. By measuring the cyclotron frequency for data with a given number of between one and ten detected ions per shot, we determined that limiting the analysis to events with five or fewer detected ions, corresponding to eight or fewer ions in the trap, minimizes this relative shift in the frequency ratio to well below 1×10^{-9}. Also, the set of cyclotron frequency ratios has a near-unity Birge ratio [20] of 1.091(51), which indicates that additional statistical effects are unlikely. These frequency ratios are shown in Fig. 3 about the average cyclotron frequency ratio, $\nu_c(^{96}\mathrm{Zr}^+)/\nu_c(^{96}\mathrm{Mo}^+) =$ 0.999 962 436 3(15).

The ^{96}Zr $0\nu\beta\beta$-decay Q value is given by

$$Q_{\beta\beta} = \left[m(^{96}\mathrm{Zr}) - m_e\right] c^2 \left[1 - \frac{\nu_c\left(^{96}\mathrm{Zr}^+\right)}{\nu_c\left(^{96}\mathrm{Mo}^+\right)}\right], \qquad (2)$$

where c is the speed of light. The new atomic mass of ^{96}Zr, $m(^{96}\mathrm{Zr})$, reported in this work, and the ratio $\nu_c(^{96}\mathrm{Zr}^+)/\nu_c(^{96}\mathrm{Mo}^+)$ results in a Q value of $Q_{\beta\beta} =$ 3355.85(15) keV.

IV. DIRECT MASS MEASUREMENTS

IV.1. ^{96}Zr direct mass measurement using $^{12}C_8$ and ^{87}Rb as references

The atomic mass value of ^{96}Zr was determined in a similar manner to the Q value measurement using the cyclotron frequency ratio between $^{96}\mathrm{Zr}^+$ and a reference species, $^{12}\mathrm{C}_8^+$. Cyclotron frequency measurements of $^{96}\mathrm{Zr}^+$ were bracketed by reference measurements of the $^{12}\mathrm{C}_8^+$ cluster, which were linearly interpolated to the time of the $^{96}\mathrm{Zr}^+$ resonance. This results in a cyclotron frequency ratio, $R = \nu_c(^{96}\mathrm{Zr}^+)/\nu_c(^{12}\mathrm{C}_8^+) = m(^{12}\mathrm{C}_8^+)/m(^{96}\mathrm{Zr}^+)$. 85 cyclotron frequency ratios were taken with approximately 2000 detected ions in each resonance.

Since the $^{12}\mathrm{C}_8$ cluster and ^{96}Zr have the same mass number, mass-dependent systematic errors, for example due to electric field imperfections [18], are much less than the relative statistical uncertainty of 1.8×10^{-9}. The relative uncertainty related to Coulomb interactions within a species was minimized to well below 1×10^{-9} by limiting the anaysis to events with five or fewer detected ions. The relative uncertainty due to nonlinear magnetic field fluctuations is again considered to be insignificant [19]. These known systematic effects are negligible compared to the statistical uncertainty. The Birge ratio is found to be 0.979(64), which indicates that the fluctuations in the data are statistical and the residual systematic error is negligible. The average cyclotron frequency ratio is measured as $\nu_c(^{96}\mathrm{Zr}^+)/\nu_c(^{12}\mathrm{C}_8^+) = 1.000\ 956\ 363\ 0(18)$.

The atomic mass of ^{96}Zr is given by

$$m(^{96}\mathrm{Zr}) = \left[8 \cdot m(^{12}\mathrm{C}) - m_e + BE(^{12}\mathrm{C}_8)\right] \times \left[\frac{\nu_c(^{96}\mathrm{Zr}^+)}{\nu_c(^{12}\mathrm{C}_8^+)}\right]^{-1} + m_e, \qquad (3)$$

where $m(^{12}\mathrm{C})$ is the atomic mass of a carbon atom and $BE(^{12}\mathrm{C}_8)$ is the molecular binding energy of the $^{12}\mathrm{C}_8$ cluster. The ionization potentials of ^{96}Zr and $^{12}\mathrm{C}_8$ cluster were not included in the calculation as they differ only by a few eV. However the molecular binding energy of the $^{12}\mathrm{C}_8$ cluster, which, in its most stable ringlike geometry, is -45.18 eV [21], is significant at our uncertainty level and was thus included in Eqn. 3. Clusters of ^{12}C are ideal reference isotopes because they define the mass standard, $m(^{12}\mathrm{C}) = 12$ u, and thus introduce no additional uncertainty. The resulting atomic mass value is $m(^{96}\mathrm{Zr}) = 95.908\ 277\ 35(17)$ u and the mass excess is $-85\ 439.11(16)$ keV.

We have also performed ^{96}Zr direct mass measurement using ^{87}Rb as the reference species. 52 cyclotron frequency ratios were taken with approximately 2500 ions in each resonance. Because the measured ^{96}Zr and the ^{87}Rb reference have different mass numbers, the measured frequency ratio is corrected to account for the mass-dependent systematic effects arising from, for example,

TABLE I. Measured frequency ratios, ν_c(ion)/ν_c(reference), calculated atomic mass and mass excess (ME) values of $^{90-92,94}$Zr using $^{12}C_8$ and ^{87}Rb references and their comparison to the values from 2012 Atomic Mass Evaluation [22]. Differences in the mass excess values, ΔME = ME$_{\rm LEBIT}$ − ME$_{\rm AME2012}$, are also listed.

Isotope	Reference	Frequency Ratio	Mass (u)	ME (keV)	AME2012 (keV)	ΔME (keV)
^{90}Zr	$^{12}C_8$	1.067 797 775 3(27)	89.904 698 48(23)	−88 772.80(21)	−88 773.6(1.8)	0.8(1.9)
	^{87}Rb	0.966 680 978 1(17)	89.904 698 94(16)	−88 772.37(15)		1.2(1.9)
^{91}Zr	$^{12}C_8$	1.056 040 423 8(29)	90.905 640 00(25)	−87 895.78(24)	−87 896.2(1.8)	0.4(1.8)
	^{87}Rb	0.956 037 010 1(20)	90.905 640 23(19)	−87 895.57(17)		0.6(1.8)
^{92}Zr	$^{12}C_8$	1.044 556 743 7(20)	91.905 035 22(17)	−88 459.13(16)	−88 459.6(1.8)	0.5(1.8)
	^{87}Rb	0.945 640 795 4(22)	91.905 035 43(21)	−88 458.94(19)		0.7(1.8)
^{94}Zr	$^{12}C_8$	1.022 295 621 2(22)	93.906 312 57(21)	−87 269.29(19)	−87 270.9(1.9)	1.6(1.9)
	^{87}Rb	0.925 487 730 3(36)	93.906 312 24(36)	−87 269.59(34)		1.3(1.9)
^{96}Zr	$^{12}C_8$	1.000 956 363 0(18)	95.908 277 35(17)	−85 439.11(16)	−85 444.6(2.0)	5.5(2.0)
	^{87}Rb	0.906 169 219 3(32)	95.908 277 80(31)	−85 438.69(29)		5.9(2.0)

Penning trap imperfections such as deviations from a purely quadrupole electric potential and trap misalignment with respect to the magnetic field. To determine the mass-dependent systematic shift at LEBIT, we have also performed measurements using ions with very well known masses, such as $^{39}K^+$, $^{85}Rb^+$, $^{133}Cs^+$ as well as several carbon clusters. From that study, the value of the mass-dependent systematic shift is determined to be 2.0×10^{-10}/u. This shift is also added quadratically to the statistical uncertainty. The average cyclotron frequency ratio is measured as $\nu_c(^{96}Zr^+)/\nu_c(^{87}Rb^+)$ = 0.906 169 219 3(33) resulting in an atomic mass value of $m(^{96}Zr)$ = 95.908 277 80(35) u and the mass excess of −85 438.69(32) keV, in agreement with the values obtained using $^{12}C_8$ as the reference. The results are shown in Table I which summarizes the mass values of the Zr isotopes determined in this work and their comparison to 2012 Atomic Mass Evaluation (AME2012) [22].

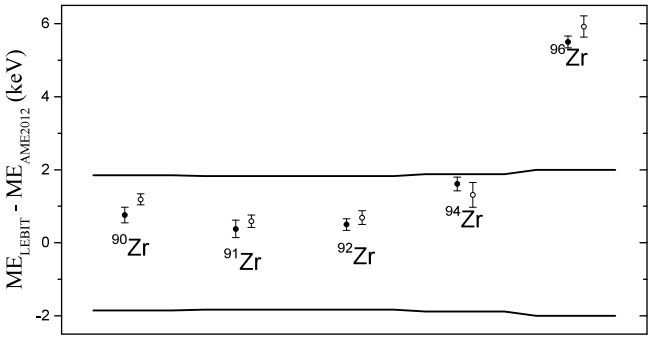

FIG. 4. Comparison of the mass excess values of the LEBIT measurements to AME2012 for $^{90-92,94,96}$Zr with $^{12}C_8^+$ (solid circles) and $^{87}Rb^+$ (open circles) used as reference species. The AME2012 uncertainty for each isotope is shown as horizontal lines around 0.

IV.2. $^{90-92,94}$Zr direct mass measurements using $^{12}C_8$ and ^{87}Rb as references

We have determined the atomic mass values of other stable $^{90-92,94}$Zr isotopes using cyclotron frequency ratio measurements similar to those described for the case of ^{96}Zr. Two different ions, $^{12}C_8^+$ and $^{87}Rb^+$, were used as reference species. The number of cyclotron frequency ratio measurements for $^{90}Zr^+$, $^{91}Zr^+$, $^{92}Zr^+$ and $^{94}Zr^+$, where $^{12}C_8^+$ is used as the reference ion, were 31, 30, 44 and 29, respectively. In the case of $^{87}Rb^+$ as the reference ion, the number of measurements were 18, 15, 15 and 15, respectively.

The frequency ratios, ν_c(ion)/ν_c(reference), corrected for mass-dependent systematic effects, our calculated atomic mass and mass excess (ME) values, and their comparison to AME2012, ΔME = ME$_{\rm LEBIT}$ − ME$_{\rm AME2012}$, are summarized in Table I. The total uncertainties of the frequency ratios range from 1.7×10^{-9} to 3.6×10^{-9}.

In the AME2012, the atomic masses of the Zr isotopes are mainly determined from neutron separation energy data from thermal neutron capture, (n,γ), reactions. Masses 90-91, 91-92, 92-93, 94-95, 96-97 are linked in this way [23]. A graphical comparison of the LEBIT mass excess values to the AME2012 values is shown in Fig. 4. The results obtained using two different reference species are in good agreement. Our results, except in the case of ^{96}Zr, fall within the uncertainty limits of AME2012.

IV.3. $^{92,94-98,100}$Mo direct mass measurements using $^{12}C_8$ and ^{87}Rb as references

Similar to the atomic mass measurements of the stable Zr isotopes, we have determined the atomic mass values for the stable Mo isotopes, $^{92,94-98,100}$Mo, with both $^{12}C_8^+$ and $^{87}Rb^+$ ions as the reference species. With $^{12}C_8^+$ as the reference ion, the number of cyclotron frequency ratio measurements were 12 for each of $^{92}Mo^+$ and $^{94}Mo^+$; and 11 for each of $^{95-98}Mo^+$ and $^{100}Mo^+$. When $^{87}Rb^+$ is used as the reference ion, the corresponding numbers were 13 for $^{92}Mo^+$ and 12 for the other Mo isotopes. The frequency ratios are corrected to account

TABLE II. Measured frequency ratios, ν_c(ion)/ν_c(reference), calculated atomic mass and mass excess (ME) values of $^{92,94-98,100}$Mo using $^{12}C_8$ and ^{87}Rb references and their comparison to the values from the 2012 Atomic Mass Evaluation [22]. Differences in the mass excess values, ΔME = ME$_{\rm LEBIT}$ − ME$_{\rm AME2012}$, are also listed.

Isotope	Reference	Frequency Ratio	Mass (u)	ME (keV)	AME2012 (keV)	ΔME (keV)
^{92}Mo	$^{12}C_8$	1.044 536 609 6(53)	91.906 806 74(46)	−86 808.97(43)	−86 807.8(0.8)	-1.2(0.9)
	^{87}Rb	0.945 622 564 3(19)	91.906 807 29(19)	−86 808.45(18)		-0.6(0.8)
^{94}Mo	$^{12}C_8$	1.022 309 000 4(46)	93.905 083 60(42)	−88 414.06(39)	−88 412.8(0.4)	-1.3(0.6)
	^{87}Rb	0.925 499 837 5(34)	93.905 083 79(35)	−88 413.89(33)		-1.1(0.6)
^{95}Mo	$^{12}C_8$	1.011 528 988 2(40)	94.905 837 97(37)	−87 711.37(35)	−87 710.6(0.4)	-0.8(0.6)
	^{87}Rb	0.915 740 660 7(42)	94.905 837 62(43)	−87 711.69(40)		-1.1(0.6)
^{96}Mo	$^{12}C_8$	1.000 993 960 8(49)	95.904 675 01(47)	−88 794.66(44)	−88 793.6(0.4)	-1.1(0.6)
	^{87}Rb	0.906 203 258 6(41)	95.904 675 26(43)	−88 794.43(40)		-0.8(0.6)
^{97}Mo	$^{12}C_8$	0.990 650 506 5(37)	96.906 017 02(36)	−87 544.59(34)	−87 543.6(0.4)	-1.0(0.6)
	^{87}Rb	0.896 839 293 3(36)	96.906 017 35(38)	−87 544.28(36)		-0.7(0.6)
^{98}Mo	$^{12}C_8$	0.980 538 209 5(53)	97.905 403 77(53)	−88 115.83(49)	−88 114.8(0.4)	-1.0(0.7)
	^{87}Rb	0.887 684 602 1(58)	97.905 403 46(64)	−88 116.12(60)		-1.3(0.8)
^{100}Mo	$^{12}C_8$	0.960 888 920 1(38)	99.907 467 52(39)	−86 193.46(36)	−86 189.5(1.0)	-3.9(1.1)
	^{87}Rb	0.869 896 017 8(50)	99.907 468 95(57)	−86 192.12(53)		-2.6(1.2)

for the mass-dependent systematic effects using the value 2.0×10^{-10}/u. The total uncertainties of the frequency ratios range from 1.9×10^{-9} to 5.8×10^{-9}.

All of the stable Mo isotope masses were recently measured at the JYFLTRAP Penning-trap mass spectrometer with a relative accuracy of 1×10^{-9} with ^{85}Rb used as the reference [24]. These measurements are the primary influence for ^{92}Mo and ^{100}Mo AME2012 values and also contribute significantly to the masses of other stable Mo isotopes [22]. The primary contribution to $^{94-98}$Mo AME2012 values are (n,γ) reaction measurements which link the masses 94-95, 95-96, 96-97, and 97-98 [23]. For the AME2012 value of ^{100}Mo mass, the ISOLTRAP mass measurement with ^{85}Rb reference [25] and the JYFLTRAP ^{100}Mo-^{100}Ru Q value measurement [26] are other contributors.

Table II summarizes the measured frequency ratios, calculated atomic mass and mass excess (ME) values and and their difference from the AME2012 values. Given in Fig. 5 is the comparison of the LEBIT mass excess values to AME2012 mass excess values also showing the agreement between the LEBIT results using two different reference species, except in the case of ^{100}Mo, where the two results deviate by more than 1-σ. Considering that the frequency measurements for all Mo isotopes were performed in one 85-hour long run for each reference mass and the difference between our results is significant only in the case of ^{100}Mo, suggests that the deviation should be attributed to statistical reasons rather than any systematic effect. Shown also in Fig. 5 is the JYFLTRAP ^{100}Mo mass excess [24] compared to the AME2012 value. Although the LEBIT results for ^{100}Mo disagree with the AME2012 value by a significant amount, they are in better agreement with the JYFLTRAP value, which is also a Penning trap measurement.

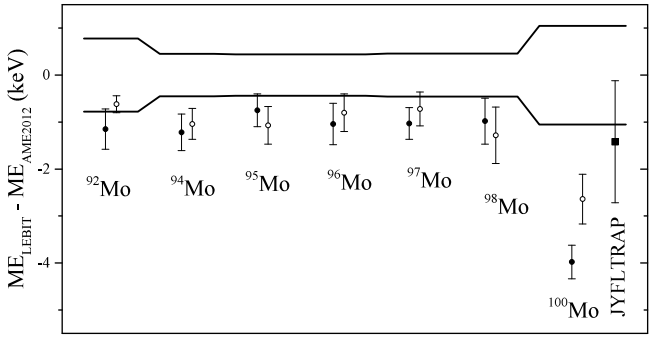

FIG. 5. Comparison of the mass excess values of the LEBIT measurements to AME2012 for $^{92,94-98,100}$Mo with $^{12}C_8^+$ (solid circles) and $^{87}Rb^+$ (open circles) used as reference species. The AME2012 uncertainty for each isotope is shown as horizontal lines around 0. The JYFLTRAP ^{100}Mo mass excess comparison is also shown.

V. Q VALUE OF ^{92}Mo AND ^{94}Zr

We have also measured the double-electron capture Q value of ^{92}Mo and the double-β decay Q value of ^{94}Zr. The method used is similar to the Q value measurement of ^{96}Zr discussed earlier. In the case of the double-electron capture Q value measurement of ^{92}Mo, we have collected 78 frequency ratios, $\nu_c(^{92}Mo^+)/\nu_c(^{92}Zr^+)$, over a period of 72 hours. Only events with 5 or fewer detected ions were included in the analysis. The frequency ratio $\nu_c(^{92}Mo^+)/\nu_c(^{92}Zr^+)$ is found to be 0.999 980 733 6(17). Using this frequency ratio and the new atomic mass of ^{92}Mo reported in Table II of this work, we obtain the double-electron capture Q value of ^{92}Mo, $Q_{\epsilon\epsilon}$ = 1649.51(15) keV. This value deviates from AME2012 value, 1651.8(2.0), by 2.4 keV, a 1.2-σ shift, and is one or-

der of magnitude more precise. For a comparison to our direct mass measurements of ^{92}Mo and ^{92}Zr with $^{12}C_8$ used as the reference, using the mass values from Tables I and II, we calculate the Q value to be 1650.16(46), which is in good agreement with our direct Q value determination.

The double-β decay Q value of ^{94}Zr is obtained from 51 $\nu_c(^{94}Zr^+)/\nu_c(^{94}Mo^+)$ frequency ratios collected over 47 hours. After a similar treatment as above, the frequency ratio, $\nu_c(^{94}Zr^+)/\nu_c(^{94}Mo^+)$, is found to be 0.999 986 915 0(23), which corresponds to $Q_{\beta\beta}$=1144.56(31) keV. Our value is higher than the AME2012 value, 1141.9(1.9), by 2.7 keV. Using our new ^{94}Zr and ^{94}Mo direct mass measurement values from Tables I and II with $^{12}C_8$ used as the reference, we obtain 1144.77(44) which is in good agreement with our direct double-β decay Q value.

VI. DISCUSSION

The new, improved ^{96}Zr double-β decay Q value, 3355.85(15) keV, deviates by nearly 7 keV from the previously accepted value, 3349.02(1.98) keV and is more precise by an order of magnitude. The previous value was derived primarily from the difference between the atomic masses of the parent and daughter species as listed in AME2012 [22]. The ^{96}Zr AME2012 mass value is principally based on the neutron separation energy from the thermal neutron capture reaction $^{96}\mathrm{Zr}\,(\mathrm{n},\gamma)^{97}\mathrm{Zr}$ [23] and the mass value for ^{97}Zr.

Our new ^{96}Zr mass excess value measured by using $^{12}C_8$ as the reference, ME(^{96}Zr) = $-$85439.11(16) keV, deviates by 5.5 keV, a nearly 3-σ shift, from AME2012 value and is one order of magnitude more precise. While the Q value obtained from Eqn. 2 uses our new mass value for ^{96}Zr, it depends primarily on the cyclotron frequency ratio, $\nu_c(^{96}Zr^+)/\nu_c(^{96}Mo^+)$, and is largely insensitive to the mass value itself. The change from the 2012 AME-based Q value is primarily due to the large shift in the ^{96}Zr mass value relative to the ^{96}Mo mass value. Using our new ^{96}Mo mass excess that is measured with $^{12}C_8$ reference, ME(^{96}Mo) = $-$88794.66(44) keV, and our new ^{96}Zr mass excess given above, one can calculate the Q value from these two separate measurements to be 3355.55(47), which is in good agreement with our direct Q value measurement.

Using the new Q value, the phase space factors for both the $2\nu\beta\beta$-decay, $G_{2\nu}$, and the $0\nu\beta\beta$-decay, $G_{0\nu}$ can be updated following the procedures outlined in Ref. [27] using a weak axial-vector coupling constant of $g_A = 1.254$. The new phase space factors and Q value are listed in Table III along with those using the AME2012 value. The new Q value is larger than the AME2012 value and increases the phase space factors, $G_{2\nu}$ by 1.8% and $G_{0\nu}$ by 0.8%. The uncertainties for both are reduced by an order of magnitude. Using the new $2\nu\beta\beta$-decay phase space factor, $G_{2\nu}$=1.9514(8)$\times 10^{-17}$ yr^{-1}, and the measured half life of $T^{2\nu}_{1/2}$=2.3(2)$\times 10^{19}$ yr [28], one calculates the $2\nu\beta\beta$-decay matrix element $|M_{2\nu}|$=0.047(2).

TABLE III. ^{96}Zr $0\nu\beta\beta$-decay Q value based on the direct measurement of $\nu_c(^{96}Zr^+)/\nu_c(^{96}Mo^+)$ along with the 2012 AME value. Corresponding $0\nu\beta\beta$-decay and $2\nu\beta\beta$-decay phase-space factors calculated using $g_A = 1.254$ and following the procedure given in [27] are also listed.

Source	Q value (keV)	$G_{2\nu}$ ($\times 10^{-17}$ yr^{-1})	$G_{0\nu}$ ($\times 10^{-14}$ yr^{-1})
LEBIT	3355.85(15)	1.9514(8)	6.0334(10)
AME2012	3349.02(1.98)	1.916(10)	5.987(13)

We have also determined the masses of $^{90-92,94}$Zr using two seperate reference species, $^{12}C_8$ and ^{87}Rb, with about an order of magnitude better precision compared to AME2012. The masses are found to be slightly less bound than those given in AME2012, however all within the uncertainty limits of AME2012. The new values will also influence the isotopes of the neighboring Y and Nb that use Zr masses.

For the stable Mo isotopes, our level of precision was about the same as the one given in AME2012. We have, however, obtained about a factor of 2 better precision in ^{92}Mo and ^{100}Mo. All the measured Mo isotopes were found to be slightly more bound compared to AME2012 except in the case of ^{100}Mo where the deviation was about -3 keV, a 3-σ shift, from the AME2012 value. The newly measured values will have an effect of on the evaluation of the isotopes of Nb and Tc. The new mass values determined using $^{12}C_8$ and ^{87}Rb as the reference species also support our other independent direct Q value measurements; i.e. the double-electron capture Q value of ^{92}Mo and the double-β decay Q value of ^{94}Zr.

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