Precise determination of the ¹¹³Cd fourth-forbidden non-unique β-decay Q value

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Using Penning trap mass spectrometry, we have performed a precise determination of the Q value for the highly forbidden β decay of ¹¹³Cd. An independent measurement of the Q value fixes the end-point energy in a fit to the ¹¹³Cd β-decay spectrum. This provides a strong test of systematics for detectors that have observed this decay, such as those developed for $\beta\beta$-decay searches in cadmium and other isotopes. It will also aid in the theoretical description of the β-decay spectrum. The result, $Q_\beta = 323.89(27)$ keV, agrees at the 1.3σ level with the value obtained from the 2012 Atomic Mass Evaluation [Chin. Phys. C **36**, 1603 (2012)], but is a factor of almost four more precise. We also report improved values for the atomic masses of ¹¹³Cd, ¹¹³In, and ¹¹²Cd.

I. INTRODUCTION

Studies of nuclear β decay have a long history in the development of atomic and nuclear theory and in the construction of the standard model of particle physics. They continue to play an important role in areas of current interest in modern physics. For example, superallowed β decay [1] and $\beta - \nu$ correlation studies [2] test the maximum parity violation assumption and V-A character of the electroweak interaction; β-decay spectroscopy experiments with ³H [3,4], ¹⁸⁷Re [5], and ¹⁶³Ho [6–8] aim for a direct determination of the electron neutrino mass with sub-eV sensitivity; and experimental searches for neutrinoless $\beta\beta$ decay could lead to the observation of lepton number violation and the determination of the Majorana or Dirac nature of the neutrino [9]. Interesting areas of study remain open for nuclear theory, such as the description of ultralow Q value β decays [10–12] and highly forbidden β decays [13–15].

¹¹³Cd is one of only three nuclei known to exist in nature for which the dominant decay process is a fourfold forbidden nonunique β decay (the others are ⁵⁰V [16,17] and ¹¹⁵In [15,18–20]). The decay scheme for ¹¹³Cd is shown in Fig. 1. The highly forbidden nature of the decay results in a long half-life of $\sim 8 \times 10^{15}$ yr. The first evidence for ¹¹³Cd β decay was reported by Greth *et al.* in 1970 [21]. The experiment used an enriched ¹¹³Cd foil and a proportional counter, and obtained a half-life of $9.3(1.9) \times 10^{15}$ yr. Since then six additional studies of ¹¹³Cd β decay have been performed [22–27]. These experiments utilized CdTe [22] and CdZnTe [25,27] semiconductor detectors, CdWO₄ low temperature bolometers [23], and CdWO₄ scintillator detectors [24,26] that have been developed for experimental searches for neutrinoless $\beta\beta$ decay in cadmium and tellurium isotopes.

The ¹¹³Cd half-life measurements obtained from Refs. [21–27] are in good agreement. However, there is a slight discrepancy between the Q values obtained from the end-point of the energy spectra. The CdTe and CdZnTe semiconductor detectors, and the CdWO₄ bolometer obtained Q values consistent with that determined from the most recent Atomic Mass Evaluation (AME2012), $Q_\beta = 322.6(1.0)$ keV [28]. Whereas the CdWO₄ scintillator detectors obtained Q values that were higher than the AME2012, but with larger systematic uncertainties: $Q_\beta = 337.4(0.3)(22)$ keV [24] and $Q_\beta = 344.9(0.2)(21)$ keV [26], where the numbers in parentheses represent the statistical and systematic uncertainties, respectively. An independent determination of the Q value will resolve this discrepancy and provide a check of systematics in these detectors. In addition, it provides an accurate Q value for the calculation of the phase space factor in the theoretical description of the β-decay spectrum.

In this paper, we report on the first direct determination of the ¹¹³Cd β-decay Q value obtained from measurements of the cyclotron frequency ratio of ¹¹³Cd⁺ and ¹¹³In⁺ ions in a Penning trap.

II. EXPERIMENTAL METHOD

A schematic of the Low-Energy Beam and Ion Trap (LEBIT) facility [29] that was used in this work is shown in Fig. 2. Cd⁺ and In⁺ ions were produced with a laser ablation ion source (LAS) [30], in which cadmium and indium foils with natural isotopic abundances were installed. In the LAS ions are extracted at an energy of 5 keV from the target foil and focused into a 90° quadrupole bender that steers the ions into a two-stage radiofrequency (rf) quadrupole beam cooler and buncher [31,32]. Low emittance ion bunches of ~ 100 ns duration are released from the cooler/buncher and transported to the LEBIT Penning trap [33]. Before entering the magnetic field the ions pass through a fast electrostatic kicker that is used as a time-of-flight gate to select ions of the desired A/q. Ions are then decelerated by a series of retardation electrodes and captured in the Penning trap.
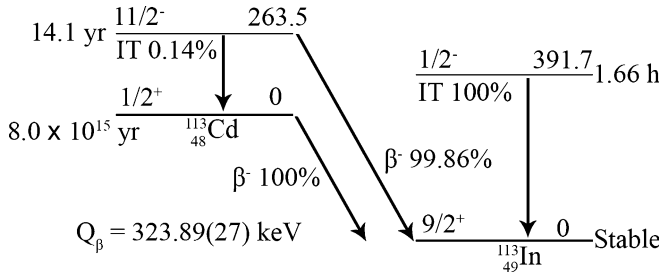
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FIG. 1. Nuclear level scheme for β-decay of ^{113}Cd.

The Penning trap itself consists of a uniform 9.4 T magnetic field produced by a horizontal bore superconducting solenoid, and a quadratic electrostatic potential produced by hyperbolic ring and end-cap electrodes. The ring electrode is segmented to allow dipole and quadrupole rf pulses to be applied to the ions. Correction ring and correction tube electrodes are included that enable higher order electric field terms to be nulled. In the Penning trap, an ion experiences three normal modes of motion: the axial mode, and the reduced-cyclotron and magnetron radial modes, with characteristic frequencies f_z, f_+, and f_-, respectively (see, e.g., Refs. [34,35] for a review of Penning trap physics).

At LEBIT, the time-of-flight ion-cyclotron-resonance (TOF-ICR) technique is used to determine the cyclotron frequency, $f_c = qB/2\pi m$, of an ion with mass-to-charge ratio m/q in the magnetic field, B. In this technique, a pulsed quadrupole rf drive is applied to the ions at a frequency f_{rf} near to the sum frequency $f_+ + f_- = f_c$, which results in a coupling of the reduced-cyclotron and magnetron modes. Full conversion of magnetron to reduced-cyclotron motion is achieved when $f_{rf} = f_c$. At LEBIT, ions are prepared in an initial magnetron orbit with radius $\sim$0.5 mm by steering them off-axis with a "Lorentz steerer" [36] before they enter the Penning trap. Contaminant ions are removed by applying a dipole rf drive at their reduced-cyclotron frequency to drive them into a large cyclotron orbit. A quadrupole rf drive is then applied to convert the initial, low frequency ($\sim$kHz) magnetron motion into high frequency ($\sim$MHz) reduced-cyclotron motion, resulting in an increase in the rotational energy of the ions. This is done using either a single rf
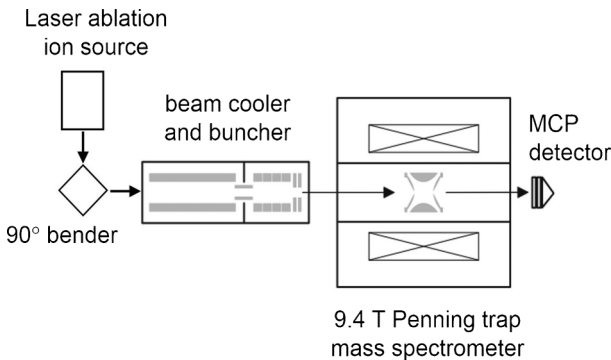

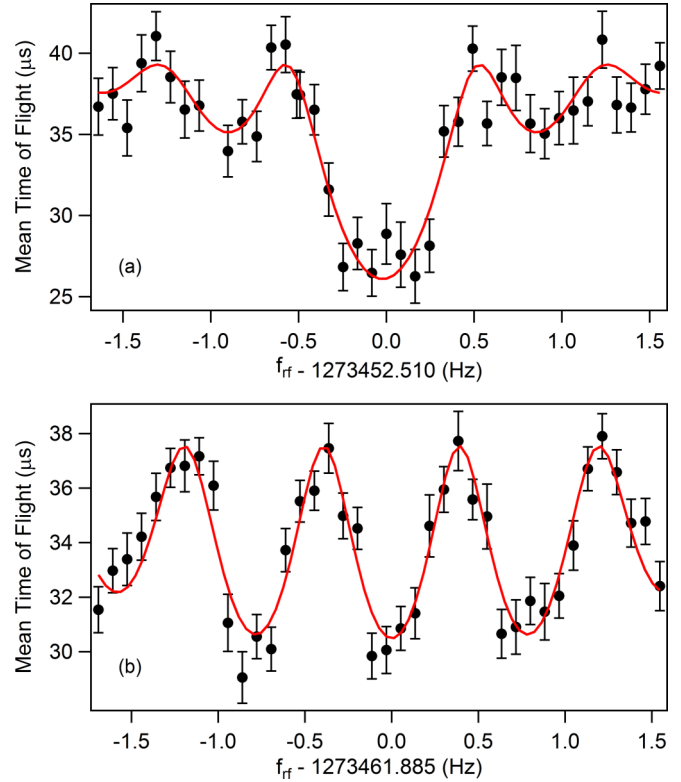FIG. 2. Schematic diagram showing the subset of components of the LEBIT facility used in this work.

FIG. 3. (a) A ^{113}Cd$^+$ time-of-flight resonance using a 1.5 s traditional quadrupole excitation scheme, and (b) a ^{113}In$^+$ time-of-flight resonance using a 1.5 s Ramsey quadrupole excitation scheme. The solid curves are fits of the theoretical traditional [38] and Ramsey [41] line shapes to the data.

pulse (traditional TOF-ICR [37,38]) or with a two-pulse sequence (Ramsey TOF-ICR [39–41]), that enables a factor of $\sim$3 increase in precision over the traditional TOF-ICR measurement in the same amount of time.

Ions are then ejected from the trap and transported through the magnetic field gradient to a microchannel plate detector (MCP), located in the fringe field of the magnet. As the ions travel through the magnetic field gradient their radial energy is converted into axial energy. Therefore, an ion's time-of-flight to the MCP depends on the amplitude of its reduced-cyclotron motion before being ejected from the trap. Hence, a sequence of measurements of time-of-flight vs f_{rf} scanned around f_c for subsequent ions bunches that are captured, probed, and ejected, results in a time-of-flight resonance curve. A representative resonance from this work for ^{113}Cd$^+$, where a 1.5 s excitation time was used, is shown in Fig. 3(a). The Ramsey excitation scheme results in an interference pattern [39–41]. A Ramsey resonance for ^{113}In$^+$ from this work, where a 300-900-300 ms on-off-on excitation pattern was used, is shown in Fig. 3(b).

In this work, the goal was to determine the ^{113}Cd β-decay Q value, defined as the energy equivalent of the mass difference between parent and daughter atoms,

$$Q_\beta = [m(^{113}\text{Cd}) - m(^{113}\text{In})]c^2. \tag{1}$$

This quantity can be obtained from the cyclotron frequency ratio of parent to daughter ions,

$$R = \frac{f_c(^{113}\text{Cd}^+)}{f_c(^{113}\text{In}^+)} = \frac{m(^{113}\text{In}^+)}{m(^{113}\text{Cd}^+)}, \quad (2)$$

in the following way:

$$Q_\beta = [(m(^{113}\text{Cd}) - m_e)c^2 + b_{\text{Cd}}](1 - R) + b_{\text{In}} - b_{\text{Cd}}, \quad (3)$$

where $b_{\text{Cd}} = 8.99\,\text{eV}$ and $b_{\text{In}} = 5.79\,\text{eV}$ are the first ionization energies of Cd and In, respectively [42], and c is the speed of light. Typically, one would determine the cyclotron frequency ratio directly by alternating between cyclotron frequency measurements on the two ion species, obtaining resonances like the ones shown in Fig. 3(a) and 3(b). Pairs of cyclotron frequency measurements for one species, e.g., the parent, are then interpolated to obtain the cyclotron frequency at the same time that the cyclotron frequency of the daughter ion was measured. In this way, magnetic field fluctuations can be eliminated to first order. The effect of nonlinear magnetic field fluctuations on the cyclotron frequency ratio was previously studied at LEBIT [43] and found to contribute at the level of $1.2(6) \times 10^{-10}$ per hour. Since the time taken to obtain a resonance was slightly less than one hour, this effect is negligible at the level of precision achieved in this work.

In this work it was not possible to alternate between ^{113}Cd and ^{113}In ions as described above. This was due to the fact that, when producing ^{113}Cd$^+$, a significant amount of ^{113}In$^+$ was also produced—presumably, after ablating the indium foil, indium was deposited on the cadmium foil and other surfaces of the LAS, so was continually ionized. It was not possible to clean the contaminant ^{113}In$^+$ ions from the trap with dipole rf cleaning. This was because the $320\,\text{keV}/c^2$ mass difference between ^{113}Cd and ^{113}In corresponds to an $\sim$4 Hz difference in the two ions' cyclotron frequencies. Hence, the ^{113}In$^+$ cleaning drive perturbed the ^{113}Cd$^+$ ions.

Instead, we measured the cyclotron frequency ratios of ^{113}In$^+$/^{115}In$^+$, ^{113}In$^+$/^{112}Cd$^+$, ^{113}Cd$^+$/^{115}In$^+$, and ^{113}Cd$^+$/^{112}Cd$^+$. These ratios were then combined in pairs to eliminate the ^{115}In$^+$ or ^{112}Cd$^+$ reference ions to obtain two independent measurements of the ^{113}Cd$^+$/^{113}In$^+$ ratio. We note that any systematic shifts in the cyclotron frequency ratios for the non-mass-doublets completely cancel when combined in pairs to obtain the ^{113}Cd$^+$/^{113}In$^+$ ratios.

After performing the ^{113}In$^+$/^{115}In$^+$ and ^{113}In$^+$/^{112}Cd$^+$ measurements, the LAS was disassembled and thoroughly cleaned, and only a (fresh) cadmium foil was installed in the LAS. However, ^{115}In$^+$ ions were still observed at approximately twice the rate as ^{113}Cd$^+$. Hence, the ^{113}Cd$^+$/^{115}In$^+$ measurement could be performed. The ^{113}In isotope, with natural abundance 4.3%, was searched for by performing a traditional TOF-ICR excitation over a frequency range that would include both the ^{113}Cd$^+$ and ^{113}In$^+$ resonances, and was determined to be present at a level of $\lesssim$10%. Nevertheless, for the cyclotron frequency measurements with ^{113}Cd$^+$, we used a 1.5 s traditional TOF-ICR scheme rather than the Ramsey TOF-ICR scheme since the resonance curves for the former scheme would be resolved for ^{113}Cd$^+$ and ^{113}In$^+$, whereas the

TABLE I. Average cyclotron frequency ratios, R, measured in this work for the ion pairs listed. N is the number of measurements used to determine the average for each ion pair. The Birge ratio for each data set is listed in the third column. The ^{113}Cd$^+$/^{113}In$^+$ ratios were obtained by combining the two ratios listed above them. The Q value was obtained from the ^{113}Cd$^+$/^{113}In$^+$ ratio using Eq. (3).

Ion pair	N	Birge ratio	R	Q value (keV)
^{113}In$^+$/^{115}In$^+$	50	1.14	1.017 712 629 0(21)	
^{113}Cd$^+$/^{115}In$^+$	33	1.06	1.017 709 498 2(31)	
^{113}Cd$^+$/^{113}In$^+$			0.999 996 923 7(37)	323.53(39)
^{113}In$^+$/^{112}Cd$^+$	18	1.17	0.991 131 399 5(28)	
^{113}Cd$^+$/^{112}Cd$^+$	47	1.02	0.991 128 343 7(25)	
^{113}Cd$^+$/^{113}In$^+$			0.999 996 916 9(38)	324.25(40)

lineshapes for the latter scheme could overlap. For all other ions in this work we used the Ramsey TOF-ICR scheme.

III. RESULTS AND DISCUSSION

The results for the average cyclotron frequency ratios that were measured in this work, along with the combined ^{113}Cd$^+$/^{113}In$^+$ ratios, and Q values determined from Eq. (3) are shown in Table I. In Eq. (3), the mass of ^{113}Cd is required to obtain the Q value. Here we have used the value for $m(^{113}$Cd$)$ obtained in this work (see Table II). However, since $m(^{113}$Cd$)$ is multiplied by the factor $(1 - R)$ in Eq. (3) the contribution of the uncertainty in $m(^{113}$Cd$)$ to the uncertainty in Q is completely negligible.

For each average ratio, between 18 and 50 pairs of TOF-ICR resonances were combined. The Birge ratio for the average cyclotron frequency ratios listed in Table I were all close to one, indicating that the statistical uncertainties describe the data reasonably well. Nevertheless, we inflated the statistical uncertainty in the average ratio by the corresponding Birge ratio [44]. We have also included small corrections for each ratio to account for systematic shifts that are introduced when ion pairs that are non-mass-doublets are compared. This systematic effect has been previously studied at LEBIT and was found to produce a shift at the level of 2.0×10^{-10} per u/e [45]. This correction was also added in quadrature to the statistical uncertainty, but is a factor of $\sim$10 smaller than our statistical uncertainty and hence provides a negligible contribution to

TABLE II. Mass excesses, ME, for ^{113}Cd, ^{113}In, and ^{112}Cd obtained from the cyclotron frequency ratios listed in Table I, and mass differences, ΔM, between our result and the AME2012 [28] values.

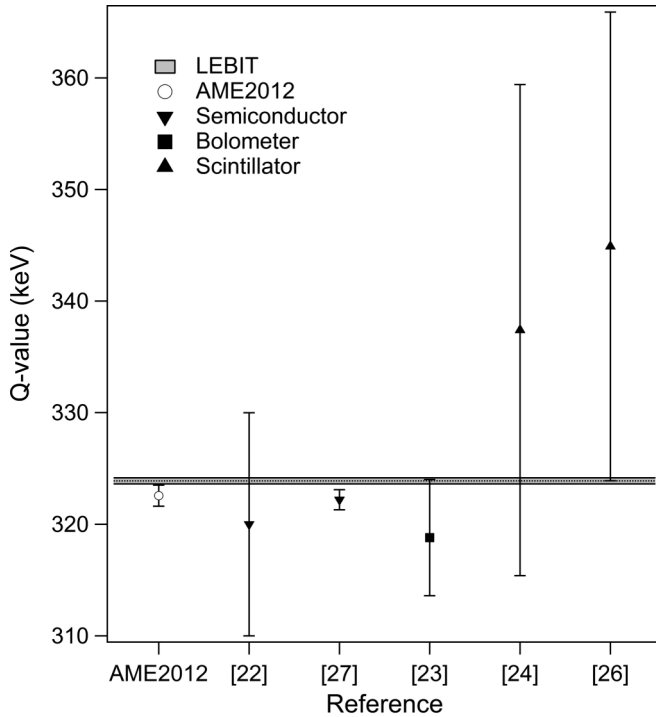| Isotope	Ref.	ME (keV/c^2)		ΔM
		This work	AME2012	(keV/c^2)
^{113}In	^{115}In	−89 366.84(22)	−89 365.82(85)	−1.02(0.88)
	^{112}Cd	−89 368.47(63)		−2.65(1.06)
^{113}Cd	^{115}In	−89 043.31(32)	−89 043.25(42)	−0.06(0.53)
	^{112}Cd	−89 044.23(62)		−0.98(0.75)
^{112}Cd	^{115}In	−90 574.51(27)	−90 575.81(56)	1.30(0.62)

FIG. 4. Result for the ^{113}Cd β-decay Q value obtained in this work (dotted line) with $\pm 1\sigma$ uncertainty (gray band). The result is compared to the value obtained from the AME2012 [28] and to measurements of the end-point of the ^{113}Cd β-decay spectrum using semiconductor detectors—Mit88 [22], Daw09 [27]; bolometers— Ale94 [23]; and scintillator detectors—Dan96 [24], Bel07 [26]. The error bars represent the statistical and any systematic uncertainties added in quadrature.

the total uncertainty. Possible additional systematic shifts due to the number of ions in the trap were made negligible by including in the analysis only data points for which the detected number of ions was $\leqslant 5$. Accounting for the 60% efficiency of our MCP, this corresponds to $\leqslant 8$ ions in the trap.

The two independent Q value measurements agree within their uncertainties and give a weighted average, $Q_\beta = 323.89(27)$ keV. This result is displayed in Fig. 4 in comparison to the Q value obtained from the AME2012 and the Q values determined using the various β-decay detectors. Our new result differs from the AME2012 value [28] by 1.3(1.0) keV and is a factor of 3.5 more precise.

The data in Table I can also be used to obtain absolute masses for ^{113}Cd and ^{113}In using ^{112}Cd and ^{115}In as references via

$$m_{\text{ion}} = (m_{\text{ref}} - m_e + b_{\text{ref}})R^{-1} + m_e - b_{\text{ion}}, \qquad (4)$$

where m_{ion} is the mass of ^{113}Cd or ^{113}In, m_{ref} is the mass of ^{112}Cd or ^{115}In, b_{ion} and b_{ref} are the first ionization energies of Cd or In, and R^{-1} is the inverse of the relevant cyclotron frequency ratio listed in Table I. The resulting mass excesses for ^{113}Cd and ^{113}In and a comparison with the values from AME2012 are listed in Table II and displayed in Fig. 5.

The uncertainties in the ^{113}In and ^{113}Cd masses obtained using the ^{112}Cd$^+$ reference ion are larger than for the ^{115}In$^+$
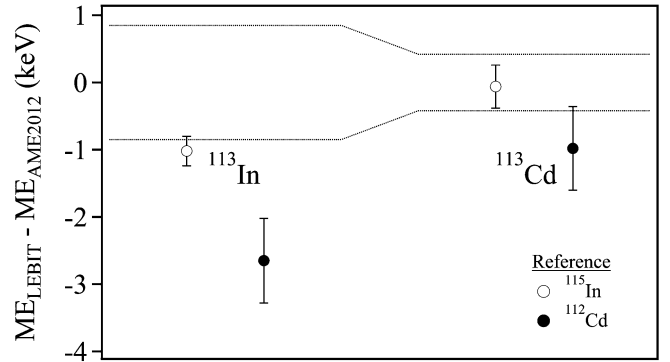

FIG. 5. Comparison between the mass excesses of ^{113}In and ^{113}Cd measured in this work (open and closed circles) and the values listed in the AME2012 [28] (the zero line). The solid lines indicate the uncertainty in the AME2012 mass values. For each isotope two independent measurements were performed using either ^{115}In or ^{112}Cd as a reference. The error bars are the uncertainties in the measured masses obtained by adding the uncertainties in the measured ratio and reference mass in quadrature.

reference ion. This is due to the 0.56 keV/c^2 uncertainty in the mass of ^{112}Cd given in the AME2012 [28] compared to the 0.012 keV/c^2 uncertainty in ^{115}In. The mass of ^{115}In was determined in a high-precision Penning trap measurement [46], whereas the mass of ^{112}Cd was determined mainly via (n,γ) and (p,d) reactions and mass spectrometry measurements that link it to ^{110}Cd, ^{110}Pd, and ^{115}Sn, which have been measured with Penning traps [46–48]. The masses we obtain using the two reference ions differ by 1.63(67) keV/c^2 for ^{113}In and 0.91(70) keV/c^2 for ^{113}Cd. We note that the systematic corrections applied to the data due to the comparison of non-mass-doublets are a factor of ~ 20 smaller than the observed mass differences and therefore cannot explain the discrepancy. Hence, these results could indicate an issue with the AME2012 value for the mass of ^{112}Cd.

By combining the four cyclotron frequency ratios in Table I to obtain two measurements of the ratio ^{112}Cd$^+$/^{115}In$^+$, we can obtain a new and more precise value for $m(^{112}$Cd$)$. From these data we obtain a weighted average for $f_c(^{112}$Cd$^+)/f_c(^{115}$In$^+) = 1.026\ 819\ 080\ 2(26)$. From this ratio, and using Eq. (4), we obtain ME(^{112}Cd$) = -90574.51(27)$ keV/c^2, which differs from the AME2012 value by 1.30(62) keV/c^2.

IV. CONCLUSION

In conclusion, we have performed a direct, high precision measurement of the Q value for the fourfold forbidden nonunique β decay of ^{113}Cd, with the result $Q_\beta = 323.89(27)$ keV. The result is in reasonably good agreement with the AME2012 value, differing by 1.3(1.0) keV, and is a factor 3.5 more precise. The result shows that the end-point energy for ^{113}Cd β decay obtained with the CdTe and CdZnTe semiconductor detectors and the CdWO$_4$ bolometer are correct, as are the values obtained with the CdWO$_4$ scintillators, which are within the estimated systematic uncertainty of these detectors—although their central value appears systematically

too large. The new Q value can be used in theoretical calculations of the ^{113}Cd half-life, and to fix the end-point in fits to the ^{113}Cd β-decay energy spectrum. We have also provided improved values for the atomic masses of ^{112,113}Cd and ^{113}In.

ACKNOWLEDGMENTS

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