Smallest Known Q Value of Any Nuclear Decay: The Rare β^- Decay of $^{115}In(9/2^+) \rightarrow {}^{115}Sn(3/2^+)$

J. S. E. Wieslander,[1,2] J. Suhonen,[2] T. Eronen,[2] M. Hult,[1,‡] V.-V. Elomaa,[2] A. Jokinen,[2] G. Marissens,[1] M. Misiaszek,[3] M. T. Mustonen,[2] S. Rahaman,[2,*] C. Weber,[2,†] and J. Äystö[2]

[1]EC-JRC-IRMM, Institute for Reference Materials and Measurements, Retieseweg 111, B-2440 Geel, Belgium
[2]Department of Physics, P.O. Box 35 (YFL), FIN-40014 University of Jyväskylä, Finland
[3]Marian Smoluchowski Institute of Physics, Jagiellonian University, ul. Reymonta 4, 30-059 Krakow, Poland

(Received 18 March 2009; published 16 September 2009)

The ground-state-to-ground-state Q_{β^-} value of ^{115}In was determined to 497.68(17) keV using a high-precision Penning trap facility at the University of Jyväskylä, Finland. From this, a Q_{β^-} value of 0.35(17) keV was obtained for the *rare* β^- *decay* to the first excited state of ^{115}Sn at 497.334(22) keV. The partial half-life was determined to $4.1(6) \times 10^{20}$ yr using ultra low-background gamma-ray spectrometry in an underground laboratory. Theoretical modeling of this 2nd-forbidden unique β^- transition was also undertaken and resulted in $Q_{\beta^-} = 57^{+19}_{-12}$ eV using the measured half-life. The discrepancy between theory and experiment could be attributed to atomic effects enhanced by the low Q value. The present study implies that this transition has the lowest Q value of any known nuclear β decay.

DOI: 10.1103/PhysRevLett.103.122501 PACS numbers: 23.40.Hc, 21.10.Dr, 23.40.Bw, 27.60.+j

From the most recent mass evaluation [1], the energy of the ground-state-to-ground-state β^- decay of ^{115}In is 499(4) keV. Given that the energy of the first excited state in ^{115}Sn is 497.334(22) keV [2], the available β^- decay energy to the excited state $^{115}Sn(3/2^+)$ is only 1.7(40) keV. It was thus uncertain whether the β^- decay $^{115}In(9/2^+) \rightarrow {}^{115}Sn(3/2^+)$ was energetically possible. However, Cattadori *et al.* [3] were first to detect this rare decay in 2005 by measuring the 497.334(22) keV γ-ray from a 929 g pure indium rod using an ultra low-background γ-ray spectrometer in the Gran Sasso National Laboratory, 3800 m water equivalent below ground. The measurement was a spin-off from experiments to characterize the bremsstrahlung spectrum of ^{115}In, as a part of the LENS neutrino project. Figure 1 shows the decay scheme of ^{115}In based on [2] with the inclusion of the β^- decay to $^{115}Sn(3/2+)$, which is the focus of this study. In the work presented here, this decay is examined through (i) measurements of the cyclotron frequencies of ^{115}In and ^{115}Sn in order to determine the ground-state-to-ground-state Q_{β^-} value of ^{115}In, which in turn is used to obtain the Q_{β^-} value for the rare decay, (ii) underground γ-ray spectrometry measurements to detect the 497.334(22) keV γ-ray from which the partial half-life of the decay is accurately determined, and (iii) a theoretical calculation of the transition energy, the Q_{β^-} value, for the rare decay.

The ground-state-to-ground-state Q_{β^-} value of ^{115}In to ^{115}Sn was measured with the Penning trap facility [4] at the accelerator laboratory of the University of Jyväskylä. The measurement procedure was similar to Q value measurements of superallowed β emitters [5]. The Q_{β^-} value was determined by a high-precision comparison of the cyclotron frequencies of the parent and daughter ions:

$$Q_{\beta^-} = m_1 - m_2 = \left(\frac{\nu_2}{\nu_1} - 1\right)(m_2 - q m_e), \tag{1}$$

where m_1, m_2, and m_e are the masses of the parent atom, the daughter atom, and the electron, $\frac{\nu_2}{\nu_1}$ is the cyclotron frequency ratio of the corresponding ions of charge state q, where a q equal to 1+ or 2+ was employed in these studies. The binding energies of the missing atomic electrons can be neglected. The ions were produced using an electric discharge ion source [6]. The composition of the discharging electrode was prepared such that an ion beam of approximately equal intensity was simultaneously produced for both ion species. A purified sample of either ^{115}Sn or ^{115}In ions was prepared using a high-resolution cleaning technique described in [7]. This was done prior to the frequency measurements of the selected species, using the time-of-flight ion-cyclotron resonance technique [8,9] in combination with radio frequency excitation applied with time-separated oscillatory fields [10].

The determination of the cyclotron frequency of one ion species lasted for approximately 1 h and a representative

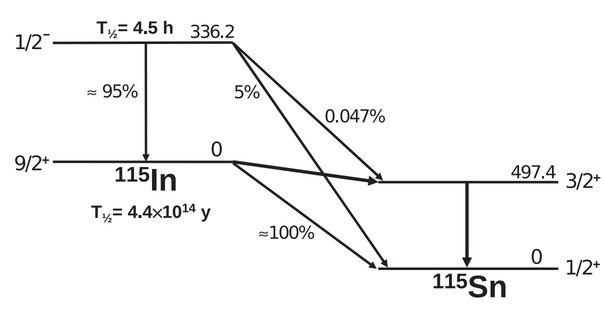

FIG. 1. Decay schemes of ^{115}In and ^{115m}In, showing the decay to the first excited state of ^{115}Sn.

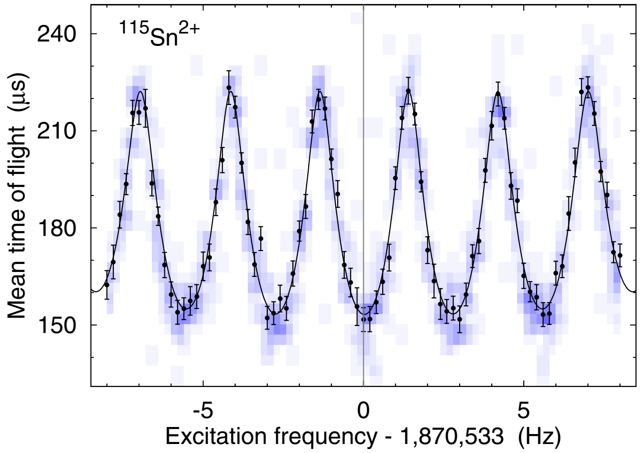

FIG. 2 (color online). A representative time-of-flight ion-cyclotron resonance obtained for $^{115}Sn^{2+}$ ions using the excitation time pattern of (50-350-50) ms (On-Off-On).

cyclotron resonance curve is shown in Fig. 2. The frequency ratio was determined from interleaving measurements of the two species, typically employed to monitor the temporal drift of the magnetic field. In this way, a weighted mean Q_{β^-} value could be calculated from several individual frequency ratios. Three sets of data were collected consisting of 11, 15, and 11 individual frequency ratios. Two of the data sets were measured with singly-charged ions while one set was measured with doubly-charged ions. The frequency ratios and Q values obtained are given in Table I. Finally, the Q_{β^-} value 0.35(17) keV of the transition $^{115}\mathrm{In}(9/2^+) \rightarrow {}^{115}\mathrm{Sn}(3/2^+)$ is deduced from the value in Table I, given that the energy of the excited state $^{115}\mathrm{Sn}(3/2^+)$ is 497.334(22) keV [2]. This indicates that the rare β^- decay is energetically possible.

Three γ-ray spectrometry measurements of a radio-pure indium sample were carried out in the underground laboratory HADES, located at a depth of 500 m water equivalent [11]. The objective was to confirm or dispute the observation of the rare decay carried out by Cattadori *et al.* [3] and to perform a dedicated measurement in a simple geometry, which would reduce the final uncertainty in comparison to their work. To detect this low level of activity it is important to perform the measurements in an underground laboratory in order to reduce the cosmogenic activation of the sample and detectors.

TABLE I. Results of the cyclotron frequency measurements. Column q denotes the charge state of the ions and column number the number of individual frequency ratios obtained.

Set	q	No.	Frequency ratio, $\frac{\nu_{\mathrm{Sn}}}{\nu_{\mathrm{In}}}$	Q_{β^-} (keV)
A	1	11	1.000 004 649 7(27)	497.66(29)
B	1	15	1.000 004 650 6(23)	497.76(25)
C	2	11	1.000 004 648 4(37)	497.52(40)
Final Q_{β^-} of ^{115}In(gs)-^{115}Sn(gs) decay				497.68(17)

The indium disc was of natural isotopic abundance with 95.71(5)% of the isotope ^{115}In [12]. After surface cleaning the disc dimensions were 10.6 cm (diameter), 4.0 cm (thickness), and 2566.13 g (total mass). Three different ultra low-background γ-ray spectrometers were made available in HADES for these measurements (measurement time in brackets): Ge-4 (48 days), Ge-7 (15 days) [13], and the sandwich spectrometer (14 days) [13].

The bremsstrahlung spectrum from the ground-state-to-ground-state β^- decay is visible in the γ-ray spectrum in Fig. 3. The excited state of ^{115}In was not present in the sample since the 336 keV γ-ray line from the main branch was not detected. The peak of interest, 497.334(22) keV, is located in a favorable energy region with a low background continuum and without interfering background peaks as displayed in Fig. 4. For Ge-4 the count rate of the continuum under the 497 keV peak was 0.38 keV^{-1} d^{-1}. Careful analysis confirmed the absence of any unknown γ-ray emitters in the spectra, which was also the conclusion of Cattadori *et al.* after a thorough study of alternative sources of the detected 497 keV γ-ray line. The full energy peak detection efficiencies were calculated using the Monte Carlo technique and the EGS4 simulation software. The geometrical model used for the Monte Carlo simulations was validated by measuring point sources (^{85}Sr, ^{137}Cs, ^{134}Cs) sandwiched between indium discs of 3, 6, and 12 mm thickness and it was confirmed that the calculated detection efficiencies agree within 3%.

The weighted mean activity of the three measurements was 0.69(9) mBq, which converts to a partial half-life of $4.1(6) \times 10^{20}$ yr. The relative combined standard uncertainty was determined to 15% and is dominated by counting statistics (14%). Other major contributions to the total uncertainty are detection efficiency (3%), isotopic abundance (0.05%), and internal conversion coefficient (0.12%). The branching ratio was calculated to $1.07(17) \times 10^{-6}$, which contains an additional uncertainty of 5.7%

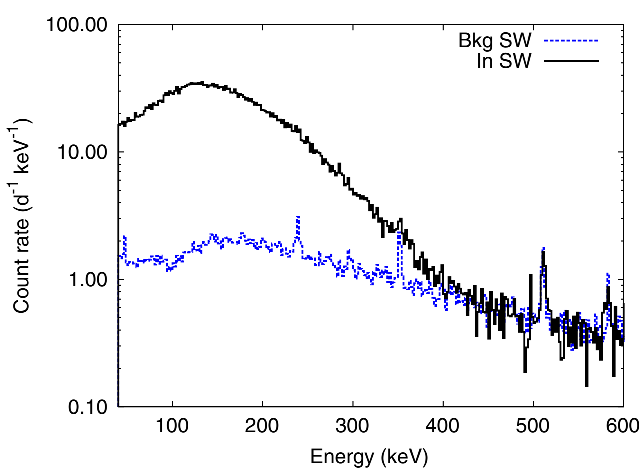

FIG. 3 (color online). γ-ray spectrum of the background (Bkg) and indium on the sandwich spectrometer (SW).

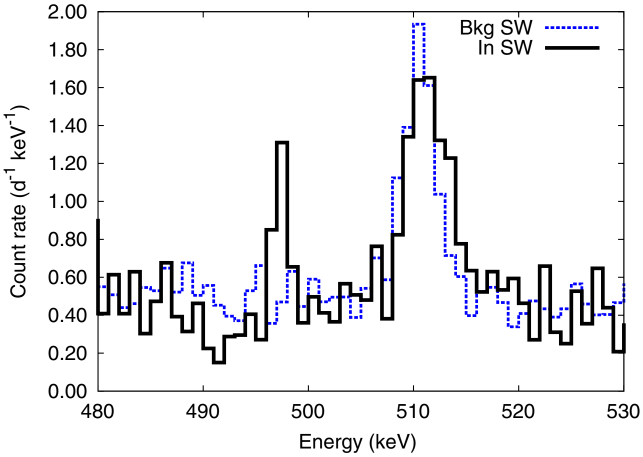

FIG. 4 (color online). γ-ray spectrum of the background (Bkg) and indium on the sandwich spectrometer (SW).

from the half-life of the ground-state-to-ground-state β^- decay. The measurements in this work agree with those of Cattadori *et al.* [3]; see Table II.

The β^- decay of the ground state of ^{115}In to the first excited state in ^{115}Sn is 2nd-forbidden unique. The half-life of a Kth forbidden unique β^- transition can be written as

$$T_{1/2} = \frac{1}{M^2 f_K(w_0, Z_f, R)}. \qquad (2)$$

Here M is the nuclear matrix element (NME) (equivalent to M_4 in Refs. [14,15] containing the nuclear-structure information). $f_K(w_0, Z_f, R)$ is a phase-space function which depends only on the end-point energy $W_0 = w_0 m_e c^2$, the charge Z_f of the daughter nucleus and the nuclear radius R. The detailed expression for the function f_K is given in a forthcoming paper.

The NME is calculated using the proton-neutron microscopic quasiparticle-phonon model (pnMQPM) where a realistic microscopic Hamiltonian is diagonalized in a basis consisting of BCS quasiparticles and their couplings with phonons obtained from the proton-neutron quasiparticle random-phase approximation. Further details are given in Ref. [16], where the model was applied to the fourth-forbidden nonunique ground-state-to-ground-state

TABLE II. Results from the γ-ray measurements on three spectrometers. The measurement by Cattadori *et al.* [3] is included for comparison. BR is the branching ratio, $t_{1/2}$ is the partial half-life, and Rel.U is the relative uncertainty.

Spectrometer	BR(10^{-6})	$t_{1/2}$(10^{20} yr)	Rel.U(%)
Ge-4	1.06(17)	4.2(7)	17
Ge-7	1.0(5)	4.4(20)	45
Sandwich	1.2(4)	3.6(12)	42
HADES final	1.07(17)	4.1(6)	15
Cattadori *et al.*	1.2(3)	3.7(10)	26

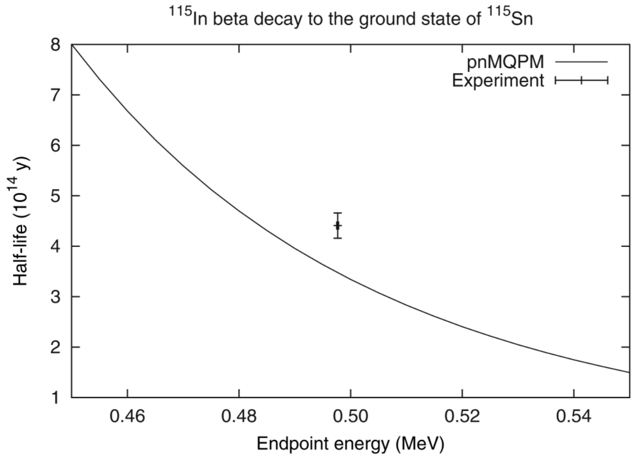

FIG. 5. Calculated half-life of the ground-state-to-ground-state β^- decay of ^{115}In as the function of the end-point energy. The experimental bars combine the half-life range from Ref. [23] and the Q value range from this Letter.

decay of ^{115}In. The theoretical curve for the half-life of that decay as a function of the Q value is shown in Fig. 5. Five percent random variations in the computed values of the NMEs easily reproduce the measured half-life so that the computed nuclear wave functions of the initial and final states seem very realistic.

Figure 6 shows the end-point energy dependence of the partial half-life for the transition ^{115}In$(9/2^+) \rightarrow$ ^{115}Sn$(3/2^+)$ by using the calculated NME and an estimated 30% uncertainty. Using the measured half-life value, $Q_{\beta^-} = 57^{+19}_{-12}$ eV is obtained. Here variations of 1 order of magnitude in the value of the NME are needed to reach the 1σ interval of measured Q_{β^-} value of Fig. 6. Such large

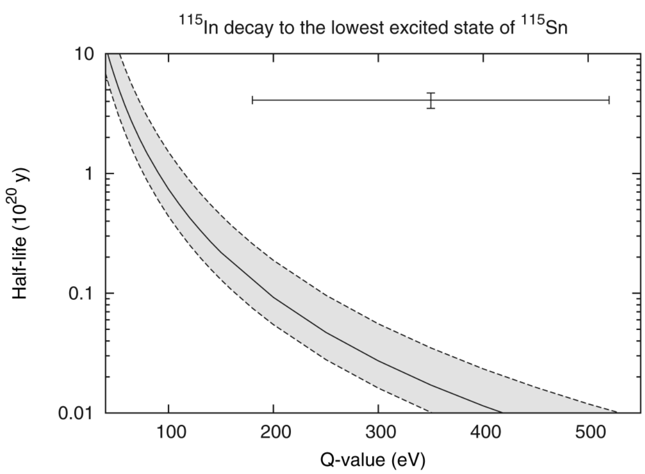

FIG. 6. Calculated relation between the Q value and partial half-life for the β^- decay ^{115}In$(9/2^+) \rightarrow$ ^{115}Sn$(3/2^+)$. The grey band indicates a conservative 30% uncertainty estimate for the NME. The experimental bars combine the half-life and Q value ranges extracted in the present underground and trap measurements.

variations in the value of the NME are unrealistic and would suggest that the excited state would have an exotic structure. A more plausible reason for the discrepancy is the exceptionally low Q value of the decay.

A small decay energy poses a challenge to the phase-space calculations. One problem is that the electron screening corrections have never been estimated for forbidden beta decays at ultralow Q values. Such estimates exist only for allowed β^- decays [17,18], showing an effect of a few percent.

A second source of correction to the phase space is the mismatch between the initial and final *atomic* states in a β decay. In [19], the associated reduction of the decay rate was calculated to be 2% for Q values larger than 20 keV. For Q values of the order of keV the formalism of [19] gives large reductions in decay rates and it breaks down completely for the ultralow Q values such as that discussed in this Letter. This correction thus shifts the shaded theory band in Fig. 6 upwards by an as yet unknown amount. Further corrections come from exchange effects [19,20] and final-state interactions [21]. In [19,20], the exchange effects were computed for Q values larger than 20 keV. The two calculations predict less than 10% effects but in opposite directions so that they contradict each other. Hence, the contribution of the exchange effects is still an open problem even for only moderately small Q values. The molecular final-state interactions were evaluated in [21] for the tritium β decay. For heavy nuclei no such evaluations exist and the related effect is a field for continuing work.

The conclusions from the present study are that the combined efforts of two very different measurements and the theoretical investigation of the β^- transition $^{115}\mathrm{In}(9/2^+) \rightarrow {}^{115}\mathrm{Sn}(3/2^+)$ suggest that this rare decay has the lowest Q value discovered for any β decay, about 1 order of magnitude smaller than that of $^{187}\mathrm{Re}$ [22]. This Letter also confirms the findings by Cattadori *et al.* [3]. Additionally, the combination of measurements and theory in this Letter clearly points to the need of further theoretical work on subtle atomic effects entering the area of theoretical modeling of β^- transitions with ultralow Q values.

This work was supported by the EU 6th Framework Programme "Integrating Infrastructure Initiative-Transnational Access," Contract No. 506065 (EURONS, JRA TRAPSPEC) and JRA 2 Contract No. 506222 (ILIAS) as well as the Finnish Center of Excellence Program 2006–2011 (Project No. 213503, Nuclear and Accelerator Based Physics Programme at JYFL).

*Present address: Physics Division P-23, Mail Stop H803, Los Alamos National Laboratory, Los Alamos, NM 87545, USA

†Present address: Fakultät für Physik, Ludwig-Maximilians-Universität München, 85748 Garching, Germany

‡mikael.hult@ec.europa.eu

[1] G. Audi, A. H. Wapstra, and C. Thibault, Nucl. Phys. **A729**, 337 (2003).
[2] J. Blachot, Nuclear Data Sheets **104**, 967 (2005).
[3] C. Cattadori *et al.*, Nucl. Phys. **A748**, 333 (2005).
[4] A. Jokinen *et al.*, Int. J. Mass Spectrom. **251**, 204 (2006).
[5] T. Eronen *et al.*, Phys. Rev. Lett. **100**, 132502 (2008).
[6] S. Rahaman *et al.*, Phys. Lett. B **662**, 111 (2008).
[7] T. Eronen *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. B **266**, 4527 (2008).
[8] M. König *et al.*, Int. J. Mass Spectrom. Ion Processes **142**, 95 (1995).
[9] G. Gräff, H. Kalinowsky, and J. Traut, Z. Phys. A **297**, 35 (1980).
[10] S. George *et al.*, Int. J. Mass Spectrom. **264**, 110 (2007).
[11] M. Hult *et al.*, Acta Chim. Slov. **53**, 1 (2006).
[12] K. J. R. Rosman and P. D. P. Taylor (IUPAC), Pure Appl. Chem. **70**, 217 (1998).
[13] J. S. E. Wieslander *et al.*, Appl. Radiat. Isot. **67**, 731 (2009).
[14] M. T. Mustonen, M. Aunola, and J. Suhonen, Phys. Rev. C **73**, 054301 (2006); Phys. Rev. C **76**, 019901(E) (2007).
[15] H. Heiskanen, M. T. Mustonen, and J. Suhonen, J. Phys. G **34**, 837 (2007).
[16] M. T. Mustonen and J. Suhonen, Phys. Lett. B **657**, 38 (2007).
[17] J. J. Matese and W. R. Johnson, Phys. Rev. **150**, 846 (1966).
[18] J. L. Lopez and L. Durand, Phys. Rev. C **37**, 535 (1988).
[19] J. N. Bahcall, Phys. Rev. **129**, 2683 (1963).
[20] M. R. Harston and N. C. Pyper, Phys. Rev. A **45**, 6282 (1992).
[21] A. Saenz and P. Froelich, Phys. Rev. C **56**, 2132 (1997).
[22] M. Galeazzi *et al.*, Phys. Rev. C **63**, 014302 (2000).
[23] L. Pfeiffer *et al.*, Phys. Rev. C **19**, 1035 (1979); L. Pfeiffer *et al.*, Phys. Rev. Lett. **41**, 63 (1978).