Determination of the 76Ge Double Beta Decay *Q* **Value**

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(Received 11 September 2000)

The Q value of the ⁷⁶Ge double beta decay has been determined by measuring the masses of ⁷⁶Ge and ⁷⁶Se in a Penning trap using neon- and fluorinelike ions. The obtained masses are 75.921 402 758(96) u and 75.919 213 795(81) u, respectively. The systematic errors of these two determinations are nearly equal, and therefore, the remaining systematic uncertainty of the *Q* value is drastically reduced. A *Q* value of 2 039.006(50) keV was obtained improving the accuracy of the accepted value by a factor of 6.

DOI: 10.1103/PhysRevLett.86.4259 PACS numbers: 21.10.Dr, 23.40.Bw, 27.50.+e, 32.10.Bi

The standard model with its many adjustable parameters has proven to be valid with an unexpectedly high precision. Many experiments have been designed in the search for a violation of this model, which if successful, may open the door to new physics. Double beta decay without emission of neutrinos would be such a violation. Already in the 1960s physicists were aware of the possibility to study the double beta decay of 76 Ge due to the attractive feature that a high purity germanium (HPGe) single crystal is a combined source and detector. These pioneer experiments were hampered by the fact that the isotopic abundance of ⁷⁶Ge is only 7.8%. Furthermore, the natural background, in particular from muons, makes conclusive measurements almost impossible in conventional laboratories. Two improvements were, therefore, essential: a very efficient shielding of the experiment and an enrichment of 76 Ge. As an example, the experiment of the Heidelberg-Moscow Collaboration [1] in the Gran Sasso tunnel fulfills these requirements. The rock amount corresponds to about 3700 m of water and the enriched abundance being 86%. In the $(0\nu \beta \beta)$ decay the electrons would appear as a peak at a position given by the *Q* value of the reaction.

In order to set a reliable limit for the $(0\nu \beta \beta)$ decay mode it is important to know the *Q* value very accurately. There are two measurements by the Manitoba group on the Q value of the double beta decay of ⁷⁶Ge using a classical mass spectrometer [2]. The first measurement [3] gave a *Q* value of 2 040.71(52) keV while a later improved measurement (less impurities) [4] gave a value of 2 038.56(32) keV. These values were obtained by comparing singly charged ions of germanium and selenium chlorides and by varying the isotopes as to get mass doublet measurements. Since the uncertainties of the two measurements do not overlap, the *Q* value has sometimes been questioned. In precision physics experience has demonstrated that it is risky to rely upon the result from only one experiment. Since Penning traps are able to deliver 1 or 2 orders of magnitude more accurate results we were motivated to check the measurements of the Manitoba group.

Our measurements are based on ion cyclotron frequency measurements. An ion with mass *m* and charge *qe* trapped in a Penning trap and moving perpendicular to a magnetic field *B* has a cyclotron frequency ν given by

$$
\nu = \frac{1}{2\pi} \frac{qeB}{m} \,. \tag{1}
$$

A frequency increase improves the mass resolving power since

$$
\frac{\nu}{\Delta \nu} = \frac{m}{\Delta m}.
$$
 (2)

One way to increase the frequency is to use a higher magnetic field (4.7 T in our case) but even more efficient is to use highly charged ions (HCI). These are supplied by the electron beam ion source (EBIS) CRYSIS at the Manne Siegbahn Laboratory and delivered to the SMILETRAP mass spectrometer [5]. CRYSIS is connected to an isotope separator that can deliver mass selected singly charged ions of practically all elements. The natural abundance of ⁷⁶Ge and ⁷⁶Se are 7.8% and 9.0%, respectively. The highly charged ions are produced by injecting singly charged ions from a plasma source (CHORDIS). Singly charged germanium ions were produced by xenon sputtering of pure germanium, whereas for selenium the ion source oven was loaded with metallic selenium. An isotopically pure beam is achieved using a magnet before injection into CRYSIS where the charge state was increased by electron bombardment. Once every second a pulse of highly charged ions (typically 10⁸ charges) was released and transported to a 90° magnet where charge selection was performed. The highly charged ions were first stopped in a retardation trap and then sent to a precision trap where the cyclotron frequencies are measured. These frequencies are determined by exciting the ion motion with a quadrupole rf field. By scanning the frequency and measuring the ion flight time to the detector it is possible to detect a resonance appearing as a flight time minimum and therefore to measure the cyclotron frequency [5,6]. In the experiment we are using an excitation time of 1 sec, which results in a resonance width close to 1 Hz, since it is a Fourier limited process.

In order to eliminate the *B* dependence we measure alternatively in a time of 1.5 min the cyclotron frequencies of both the highly charged ion and a reference ion. The molecular H_2^+ ion, which can be produced by rest gas electron-impact ionization, has been used as mass reference.

The mass of the ion is then deduced from the observed frequency ratios (Table I):

$$
R = \frac{\nu_1}{\nu_2} = \frac{q_1 m_2}{q_2 m_1},\tag{3}
$$

where the highly charged ion and the reference ion are denoted with subscripts 1 and 2, respectively. It should be noted that the observable is a frequency ratio. This means that if the two ion species are subject to the same experimental procedure the systematic errors cancel to a great extent. To deduce the mass of the neutral atom one has to correct for the mass $q_1 m_e$ of the missing electrons and their atomic binding energies *EB*:

$$
M = \frac{1}{R} \frac{q_1}{q_2} m_2 + q_1 m_e - \frac{E_B}{c^2}.
$$
 (4)

As a consistency check two charge states of 76 Ge and ⁷⁶Se were measured. We have chosen to use ${}^{76}Ge^{22+,23+}$ and ${}^{76}Se^{24+,25+}$ since these ions are Ne-like and F-like, for which it is possible to calculate the electron binding energies within a 20 eV uncertainty [7]. The binding energies used in this work are 7 934.47 and 10 111.27 eV for 76 Ge^{22+,23+}; 10 070.76 eV and 12 607.84 eV for 76 Se^{24+,25+}. The mass of H_2^+ can be calculated very accurately from the proton mass [8], the hydrogen ionization energy, the molecular binding energy of H_2^+ , and the average molecular vibrational energy [9]. The value 2.015 101 497 03(27) u has been used here.

The size and limits of possible systematic errors have been investigated using highly charged ions of ${}^{12}C$, ${}^{14}N$, ¹⁶O, ²⁰Ne, ²⁸Si, ⁴⁰Ar, and singly charged H⁺, H₂⁺, H₃⁺ all with accurately known masses. The analysis of these measurements reveals the presence of four main systematic errors. The first one is a shift that depends on the kinetic energy of the ion motion, thus being a relativistic effect. The second one is a change of the observed frequency that depends on the number of simultaneously stored ions in the trap. A third effect is a shift depending on the q/A

TABLE I. The observed frequency ratios with statistical uncertainties. (HCI: highly charged ion).

Ion	$\nu(HCI)/\nu(H_2^+)$ $\lceil \times 10^{-9} \rceil$	Statistical uncertainty [ppb]
76 Ge ²²⁺	584 015 503.986	0.71
76 Ge $^{23+}$	610 566 057.511	1.02
76 Se ²⁴⁺	637 135 385.121	0.50
76 Se $^{25+}$	663 687 465.636	0.54

ratio of the observed ion. Finally, it has been observed that the presence of a *large* amount of contaminant ions in the trap can produce a large shift of the resonance.

Relativistic mass increase.—We do not cool the ions, but rely on a sequence of selection procedures and a technique to measure the kinetic energies [5]. The initial, intermediate, and final kinetic energies are determined by a retardation technique. The average kinetic energies are \le 1.5 eV per charge implying a relativistic mass increase lower than 0.5 ppb for ${}^{76}Se^{25+}$. Since the measurement always is a comparison between the frequencies of two species, the relativistic mass increase for the involved species partly cancels, and we obtain net corrections smaller than 0.2 ppb. As a very conservative estimate of the relative uncertainty of the average kinetic energy determination, we use 50% of the correction but not less than 0.1 ppb.

Ion number dependence.—In the off-line analysis we are able to determine how many ions were trapped at a given time. By constructing resonances built on different numbers of ions we are able to conclude that the observed frequency is reduced by an amount proportional to the total charge stored in the trap during observation. From a large number of high statistics runs we can draw the following conclusions. For singly charged ions this effect is $0.127(14)$ ppb per observed charge, while for highly charged ions $(q > 20)$ the observed effect is reduced to $-0.016(10)$ ppb per observed charge. The difference between these two sets of data is at least partly (maybe fully) explained by the differences in detection efficiency. Data from singly charged ions contain more nondetected ions, thus the effect per observed charge is larger. As a consequence, in the analysis we use data with only 1 and 2 ions simultaneously stored in the trap for highly charged ions, while for H_2^+ we use 1 to 3 ions. The resulting corrections to the observed frequency of the highly charged ion are close to -0.30 ppb. However, this effect is again partly compensated by the number dependence correction of H_2^+ which ranges from -0.22 to -0.25 ppb. Therefore the net corrections are smaller than 0.05 ppb. Nevertheless, as a conservative estimate we, at present, adopt a 0.25 ppb uncertainty for the number dependence correction.

qA effect.—In the analysis of all our measurements, involving highly charged ions with well-known masses (see above) and H_2^+ ions we cannot exclude that the observed H_2^+ mass depends on the q/A ratio of the highly charged ion. For a q/A ratio difference of 0.2 (⁷⁶Ge²³⁺ - H_2^+), the effect corresponds to $-1.03(43)$ ppb. However, a test measurement using H_2^+ and H_3^+ (q/A difference = 0.17) gave a result of $+0.2(5)$ ppb, indicating a smaller (possibly zero) dependence. At present, we choose not to perform the correction, but await the accumulation of further data. However, as a conservative estimate of this possible uncertainty we take the larger value as the uncertainty.

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Uncertainty	$^{76}Ge^{22+}$	$^{76}Ge^{23+}$	76 Se ²⁴⁺	76 Se ²⁵⁺		
Reference mass	0.14	0.14	0.14	0.14		
Electron binding energies	0.26	0.26	0.26	0.26		
Average vibrational energy (H_2^+)	0.10	0.10	0.10	0.10		
Relativistic mass	0.10	0.10	0.10	0.10		
Ion number dependence	0.25	0.25	0.25	0.25		
q/A asymmetry	1.03	0.97	0.88	0.84		
Contaminant ions	0.30	0.30	0.30	0.30		
Magnetic field drift ^a	0.06	0.06	0.01	0.01		
Total systematic uncertainties	1.15	1.09	1.02	0.98		

TABLE II. Individual systematic uncertainties (the uncertainties are summed quadratically).

^aA new magnetic field stabilization system has been implemented for the selenium measurement.

Contaminant ions.—Finally, contaminant ions can be injected from the ion source or created in charge exchange processes in the trap system. The second possibility appears to be much more likely. In the case of ${}^{12}C^{6+}$ it was found that a 30% 4 He²⁺ contamination gives rise to a shift of 6 ppb. The pressure in the precision and the retardation trap has been measured to be $\leq 7.5 \times 10^{-12}$ mbar and $\leq 10^{-10}$ mbar, respectively. Therefore we estimate that the time-average contaminant rate is $\leq 3\%$. On the other hand, we consider events with only 1 and 2 highly charged ions stored in the trap. Since contaminant influence of the observations requires at least two ions, half of the measurements are not affected by this effect. Consequently, we assume the systematic uncertainty due to contaminant ions to be < 0.3 ppb.

Other uncertainties.—The influence of the uncertainty in the electron mass on the mass of an ion is $\leq 10^{-12}$ and can be ignored at the current level of precision. The uncertainty due to varying magnetic field can be estimated from the observed magnetic field drift and the time between the measurement of the two ion species. In all runs this uncertainty is well below 0.1 ppb.

Each charge state corresponds to about 15 h of data acquisition. The different runs were obtained under identical experimental conditions. As can be seen in Table I the statistical uncertainty varies from 0.50 to 1.02 ppb, while the total systematic uncertainties are below 1.15 ppb (Table II). This relatively large uncertainty originates from comparisons between two relatively different species: a singly charged light ion ($q/A = 0.5$, H_2^+) and a highly charged heavy ion $(q/A \sim 0.31, Ge^{22^+,23^+}Se^{24^+,25^+})$. However, in the *Q*-value determination the systematic uncertainties due to H_2^+ cancel out and we are left with uncertainties in two very similar species both with the same mass and similar charge states. Thus the systematic uncertainties cancel to a high degree.

From the observed ratios and the known reference mass it is possible to use Eqs. (3) and (4) to extract the neutral ⁷⁶Ge and ⁷⁶Se masses. In Table III the corrected neutral masses, both total and as obtained for the individual charge state, are presented. Note that none of the corrections are larger than the systematic uncertainties attributed to the corresponding measurement.

In conclusion, we obtained the following masses: $M(^{76}Ge) = 75.921\,402\,758(96)$) u and $M(^{76}Se)$ = 75.919 213 795(81) u. These values should be compared to the previous determinations [10], $M(^{76}Ge)$ = $75.921\,402\,7(16)$ u and $M(^{76}$ Se) = $75.919\,214\,1(16)$ u. Our mass values are 17-fold improvements of the previously best known mass values of these species.

Using the conversion factor $1 u =$ 931.494 013(37) MeV [11] we obtain a *Q* value of 2 039.006(50) keV by subtracting the two mass values. The systematic uncertainties in the masses of the two ion species are very similar. Therefore, the uncertainty in the *Q* value is drastically reduced. Our *Q* value

TABLE III. Corrected mass of ⁷⁶Ge and ⁷⁶Se determined using two different charge states. The total uncertainty on the mass determination is the quadratic sum of the statistical and systematic uncertainties.

	Corrected mass	Statistical uncertainty	Systematic uncertainty
	u	[ppb]	[ppb]
Mass of ⁷⁶ Ge from ⁷⁶ Ge ²²⁺	75, 921 402 791 670	0.71	1.15
Mass of ⁷⁶ Ge from ${}^{76}Ge^{23+}$	75.921 402 688 798	1.02	1.09
Weighted average (total uncertainty	75.921 402 758(96)		
Mass of 76 Se from 76 Se ²⁴⁺	75.919 213 800 480	0.50	1.02
Mass of 76 Se from 76 Se ²⁵⁺	75.919 213 789 104	0.54	0.98
Weighted average (total uncertainty)	75.919213795(81)		

FIG. 1. ⁷⁶Ge double beta decay *Q*-value determinations. (1) Deduced from Audi *et al.* [10]; (2) Ellis *et al.* [3]; (3) Hykawy *et al.* [4]; (4) this work.

determination confirms the last Manitoba determination 2 038.56(32) keV [4] but is a sixfold accuracy improvement (see Fig. 1) that may be useful for future experiments with higher detector resolution. Thus the new *Q* value can be used with great confidence in the search for neutrinoless double beta decay.

We gratefully acknowledge the support from the Knut and Alice Wallenberg Foundation, the Carl Trygger Foundation, The Bank of Sweden Tercentenary Foundation, The Swedish Natural Research Council, the European Community Science, and TMR Programs. We are very much indebted to E. Lindroth and P. Indelicato for informing us about their binding energy calculations prior to publication.

- [1] L. Baudis *et al.,* Phys. Rev. Lett. **83**, 41 (1999).
- [2] R. C. Barber *et al.,* Rev. Sci. Instrum. **42**, 1 (1971).
- [3] R. J. Ellis *et al.,* Nucl. Phys. **A435**, 34 (1985).
- [4] J. G. Hykawy *et al.,* Phys. Rev. Lett. **67**, 1708 (1991).
- [5] C. Carlberg *et al.,* Phys. Scr. **T73**, 347 (1997).
- [6] G. Bollen *et al.,* Nucl. Instrum. Methods Phys. Res., Sect. A **368**, 675 (1996).
- [7] G. C Rodrigues *et al.,* Phys. Rev. A **63**, 012510 (2000).
- [8] R. S. Van Dyck, Jr. *et al.,* in *Trapped Charged Particles and Fundamental Physics,* edited by D. H. E. Dublin, AIP Conf. Proc. No. 457 (AIP, New York, 1988), p. 101.
- [9] Y. Weijun, R. Alheit, and G. Werth, Z. Phys. D **28**, 87 (1993).
- [10] G. Audi and A. H. Wapstra, Nucl. Phys. A **595**, 409 (1995).
- [11] P. J. Mohr and B. N. Taylor, Rev. Mod. Phys. **72**, 351 (2000).