Masses of ^{32}Ar and ^{33}Ar for Fundamental Tests

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Masses of the short-lived radionuclides ^{32}Ar ($T_{1/2} = 98$ ms) and ^{33}Ar ($T_{1/2} = 173$ ms) have been determined with the Penning trap mass spectrometer ISOLTRAP. Relative uncertainties of 6.0×10^{-8} ($\delta m = 1.8$ keV) and 1.4×10^{-8} ($\delta m = 0.44$ keV), respectively, have been achieved. At present, these new mass data serve as the most stringent test of the quadratic form of the isobaric-multiplet mass equation. Furthermore, the improved accuracy for the mass of ^{32}Ar will allow for a better constraint on scalar contributions to the weak interaction. New mass values have also been measured for ^{44}Ar and ^{45}Ar, and a 20σ deviation for ^{44}Ar from the literature value was found and interpreted.

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Very precise mass values for specific unstable nuclides are important to test symmetry concepts in nuclear physics and to uncover physics beyond the standard model of particle interaction. Unstable rare gas isotopes are, for instance, particularly suited for beta-neutrino angular correlation measurements for which it is generally necessary to know the relevant transition energies with sufficient precision. In this Letter, we report accurate high-precision mass measurements on the short-lived isotopes ^{32,33,44,45}Ar, performed with the ISOLTRAP mass spectrometer at ISOLDE/CERN.

Wigner [1] and Weinberg and Treiman [2] noted that masses of the members of an isospin multiplet should follow the relationship

$$m(T_Z) = c_0 + c_1 T_Z + c_2 T_Z^2, \qquad (1)$$

where $T_Z = (N - Z)/2$ is the isospin projection. Since then, many measurements testing this isobaric-multiplet mass equation (IMME) have been reported [3]. In addition to the fundamental importance of isospin symmetry in nuclear physics, a practical reason for continued IMME testing is that it is widely used to predict unmeasured masses and nuclear level energies, e.g., for the mapping of the proton drip line over a wide mass range, important for determining the *rp*-process path [4].

A mass measurement on ^{33}Ar ($T = 3/2$ quartet) performed previously with ISOLTRAP indicated a possible breakdown of IMME [5]. A subsequent reevaluation of the ^{33}Cl multiplet member's mass from resonances in the ^{32}S (p, p) reaction [6] led to the revalidation of IMME. The work reported here provides improved mass values for ^{33}Ar and ^{32}Ar ($T = 2$ quintet), and these are used for further refinement and testing of IMME.

Beta-neutrino correlation measurements are a key to searches for physics beyond the standard model. For example, in pure Fermi transitions, a beta-neutrino correlation coefficient $a < 1$ would imply the presence of scalar currents [7]. Many experiments have been performed along this line, the most precise being the study of ^{32}Ar β^+ decay [8]. An essential input for the evaluation of a in [8] is the ^{32}Ar mass; however, this was known with insufficient accuracy. Therefore, IMME was used to derive a mass value with better precision, but at the expense of obtaining an a value that is no longer a purely experimental result. The experiment reported here yields a mass uncertainty for ^{32}Ar that is sufficient for a reevaluation of the beta-neutrino correlation data to obtain a new value for a that no longer relies on IMME.

The measurements on ^{32,33,44,45}Ar were performed using the Penning trap mass spectrometer ISOLTRAP [9,10] installed at the on-line mass separator facility ISOLDE/CERN [11]. Recently, the performance of ISOLTRAP has been considerably enhanced. The efficiency of the apparatus has been increased such that nuclei produced at rates of 100 ions/s and with half-lives down to ≈ 50 ms [12,13] are now accessible. Systematic tests with a carbon cluster ion source [14] have also shown that the present accuracy limit is $\delta m/m = 8 \times 10^{-9}$. Major challenges in the measurements reported here were the short half-lives [$T_{1/2}(^{32}\mathrm{Ar}) = 98$ ms and $T_{1/2}(^{33}\mathrm{Ar}) = 173$ ms], and the low ^{32}Ar production yield of ≈ 100 ions/s accompanied by an 8 orders of magnitude larger background of isobaric $^{16}\mathrm{O}_2^+$.

The argon isotopes were produced by bombarding a heated CaO target with 1.4-GeV proton pulses from the CERN proton-synchrotron-booster accelerator. A plasma ion source with a water cooled transfer line was used to suppress less volatile elements. Ions extracted from the source were accelerated to 60 keV and mass separated in ISOLDE's high-resolution mass separator. For mass 32, the resolving power of about 6000 helped to suppress the large $^{16}\mathrm{O}_2^+$ current. For short-lived nuclides, additional selection was achieved by applying a short gate to the ISOLDE beam, allowing transmission only during a period shortly after proton impact in which the radioactive

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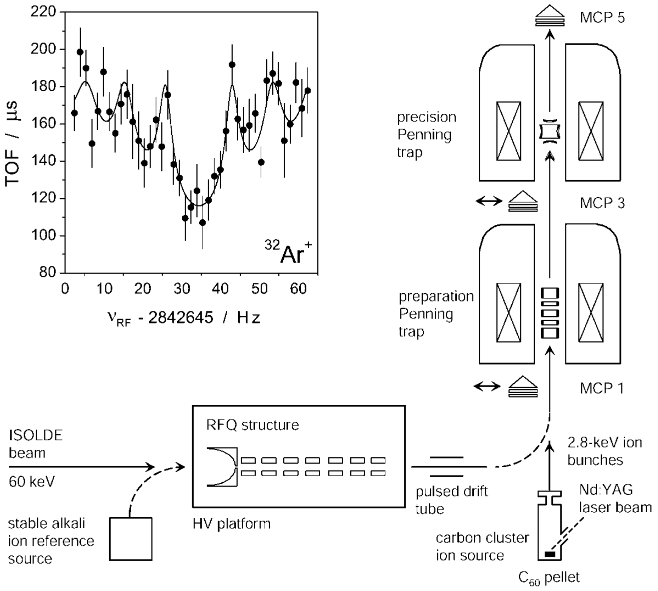

FIG. 1. ISOLTRAP mass spectrometer used for Ar isotope mass determination. Micro-channel-plate (MCP) detectors monitor ion transfer as well as record the time-of-flight resonance (MCP 5) for cyclotron frequency determination. The inset shows the cyclotron resonance of $^{32}\mathrm{Ar}^+$ with the fit of a theoretically expected curve [15].

ion production is maximum. For the $^{32}\mathrm{Ar}^+$ and $^{33}\mathrm{Ar}^+$ measurements reported here, a 10–20 ms gate was used about 70 ms after the proton pulse. Combined with the mass resolution, this time resolution reduced the O_2^+ arriving at ISOLTRAP by about 5 orders of magnitude while the number of Ar ions is reduced only by about a factor of 2–3.

As shown in Fig. 1, ISOLTRAP has three functional parts: a radio frequency quadrupole (RFQ) ion trap for ion accumulation and bunching [10] and two Penning traps: one for ion cooling and purification, and the other for mass measurement via cyclotron frequency determination [9]. The ISOLDE beam is electrostatically retarded to about 100 eV and stopped in the gas-filled linear RFQ ion trap, where ions are accumulated and cooled by energy loss in collisions with ≈ 0.5 Pa helium buffer gas. After an accumulation time of a few ms, the cooled ion bunch is ejected with a temporal width of less than 1 µs and passed through a pulsed drift tube in which the potential is reduced to ground. The ions are then stored up to several 10 ms in the preparation Penning trap where unwanted ions are removed by mass-selective buffer gas cooling [16]. In the case of $^{32}\mathrm{Ar}^+$, the $^{16}\mathrm{O}_2^+$ background was typically reduced by a factor of 10^2–10^3 without any significant losses of $^{32}\mathrm{Ar}^+$. An ion bunch is then extracted and transferred to the second Penning trap for determination of the cyclotron frequency. This entails capture of the ion bunch, excitation of ion motion with an azimuthal quadrupole rf field, followed by axial ejection of the ions and time-of-flight (TOF) analysis of the absorbed energy [17]. These steps are repeated for different trial rf frequencies near the expected cyclotron frequency. Plotting measured TOF as a function of the frequency leads to a resonance curve with a minimum in the TOF at the ions' cyclotron frequency. An example for $^{32}\mathrm{Ar}^+$ is shown in the inset of Fig. 1. A fit of the resonance curve to the theoretical function [15] yields the cyclotron frequency $\nu_c = 1/2\pi \cdot q/m \cdot B$ for an ion with mass m and charge q. The magnetic field strength B was determined via measurement of the $^{39}\mathrm{K}^+$ cyclotron frequency for which the mass is well known. Such calibration measurements were performed both before and after measurement of the cyclotron frequency for the ion of interest. The rf excitation times of $T_{\mathrm{rf}} = 0.1$ s and 0.3 s were used for ^{32}Ar and ^{33}Ar, respectively. This yielded a linewidth $\Delta\nu_c(\mathrm{FWHM}) \approx 0.9/T_{\mathrm{rf}}$ of 9 and 3 Hz and a resolving power $R = \nu_c/\Delta\nu_c(\mathrm{FWHM})$ of 3×10^5 and 1×10^6. For stable ^{36}Ar and the longer-lived ^{44}Ar ($T_{1/2} = 11.87$ min) and ^{45}Ar ($T_{1/2} = 21.48$ s), an excitation time of 1.2 s resulted in $R \approx 3 \times 10^6$. For ^{32}Ar, additional mass-selective cleaning by dipolar excitation at the reduced cyclotron frequency was necessary to remove remaining $^{16}\mathrm{O}_2^+$ ions. The total ISOLDE beam time required to perform the measurements reported here was 60 h.

Table I gives the frequency ratios and their uncertainties for the investigated argon isotopes. A detailed description of the evaluation procedure and residual systematic errors can be found in [14]. Uncertainties given in the table include the statistical uncertainty of each measurement and a relative systematic uncertainty of 8×10^{-9} added in quadrature. The atomic mass values are calculated from

$$m = (\nu_c^{\mathrm{ref}}/\nu_c)(m_{\mathrm{ref}} - m_e) + m_e, \qquad (2)$$

TABLE I. Frequency ratios relative to $^{39}\mathrm{K}^+$ and mass excesses D for ^{32,33,44,45}Ar as determined in this work. For comparison, literature values are given from the Atomic Mass Evaluation [18,19].

Nucleus	$T_{1/2}$	Frequency ratio $\nu_c^{\mathrm{ref}}/\nu_c$	D_{exp}[a]/keV	D_{lit}/keV	Difference/ keV
^{32}Ar	98 ms	0.821 213 960 4(486)	−2200.2(1.8)	−2179(50)	21.2
^{33}Ar	173 ms	0.846 681 294 3(109)	−9384.08(44)	−9382.0(4.2)	2.1
^{36}Ar	Stable	0.923 102 705 2(95)	−30 231.36(40)	−30 231.540(14)	0.18
^{44}Ar	11.87 min	1.128 357 598 2(436)	−32 672.9(1.6)	−32 262(20)	411
^{45}Ar	21.48 s	1.154 102 839 0(142)	−29 770.50(58)	−29 720(60)	50.5

[a] Using $D(^{39}\mathrm{K}) = -33\,806.94(23)$ keV [18].

TABLE II. Mass excess D in keV of the $T = 3/2$, $J^{\pi} = 1/2^{+}$ quartet for $A = 33$. Experiment 1999 gives experimental values from [5] and experiment 2002 includes new mass values from [6], [18], and this work. IMME 2002 and χ^2_ν result from a fit of the quadratic IMME, as in Eq. (1), to the data, while c_3 is the additional coefficient if IMME is assumed to be cubic.

Nucleus	T_Z	Experiment 1999	Experiment 2002	IMME 2002
^{33}P	+3/2	−26 337.7(1.1)	−26 337.7(1.1)	−26 337.6(1.0)
^{33}S	+1/2	−21 106.14(41)	−21 106.54(15)	−21 106.55(15)
^{33}Cl	−1/2	−15 460.1(1.0)	−15 455.6(8)	−15 455.39(37)
^{33}Ar	−3/2	−9381.9(4.2)	−9384.08(44)	−9384.10(43)
χ^2_ν	⋯	10.6	0.1	⋯
c_3	⋯	−2.95(90)	0.13(45)	⋯

with the electron mass m_e and the mass of the reference nuclide $m_{\rm ref}$. The resulting mass excesses ($D = m - A$u) and literature values [18,19] are also given in Table I.

The precision for all radionuclides was improved by at least 1 order of magnitude while the value for stable ^{36}Ar is already known with an uncertainty of 14 eV. All values agree very well with the literature values with the exception of ^{44}Ar, which shows a 20σ deviation. Careful examination of the ^{48}Ca(^{3}He, ^{7}Be)^{44}Ar reaction data [20] suggests that the gamma line assigned to the ^{7}Be ground state actually terminates in the 429 keV excited nuclear state [21,22], while the ground state line is hidden beneath the ^{12}C 4440 keV line. Corresponding correction of mass yields $D = -32\,691(20)$ keV, which is now in good agreement with the more precise value from this work.

The results for ^{32}Ar and ^{33}Ar can be used as a test of IMME with a precision never obtained before. For this purpose, Tables II and III summarize the measured mass excess values of all ground states of the $T = 3/2$ quartet in $A = 33$ and the $T = 2$ quintet in $A = 32$, respectively. The verification of IMME in these two cases was previously limited by the uncertainty of the Ar masses, especially for ^{32}Ar with an uncertainty of 50 keV. The last column gives the calculated mass excesses from a fit of the quadratic form of IMME to the measured values. Recently, Pyle *et al.* [6] remeasured the energy of the $J^{\pi} = 1/2^{+}$, $T = 3/2$ resonance in ^{32}S by proton scattering and found it to be about 5 keV higher than the previously accepted value [23]. This led to the change in the ^{32}Cl and ^{33}Cl masses from the 1999 values. As can be seen in the tables, the mass excesses calculated from IMME are now in excellent agreement with the experimental values.

To test IMME, one allows an additional cubic term $c_3 T_Z^3$ in Eq. (1) that should fit with $c_3 = 0$ if the quadratic form of IMME is correct. The results are given in the last line of Tables II and III and in Fig. 2 together with the c_3 coefficients for all completely measured quartets and quintets [3] including the results of this work. The accuracy achieved in these measurements on $T = 3/2, A = 33$ and $T = 2$, $A = 32$ is shown on expanded scale in the inset of Fig. 2. In both cases, the c_3 coefficient is consistent with zero within the error bars. The two investigated multiplets represent now the most stringent test of IMME.

In addition to serving for a better IMME test, the improvement in the ^{32}Ar mass can also be used to improve constraints on scalar currents. The best known value for the beta-neutrino correlation coefficient a was determined in the ^{32}Ar decay experiment performed by Adelberger *et al.* [8]. At that time, the ^{32}Ar mass was known with an uncertainty of 50 keV and consequently limited knowledge of the decay energy Q. With a dependence $\partial a/\partial Q = -1.2 \times 10^{-3}$ keV^{-1}, the mass uncertainty would have limited the accuracy for determining a to 6%. Therefore, an IMME prediction was used instead, yielding $Q = 3c_2 - c_1 = 6087.3(2.2)$ keV. With this value, a beta-neutrino correlation coefficient $a = 0.9989 \pm 0.0052({\rm stat}) \pm 0.0039({\rm syst})$ 68% C.L. was obtained, where it was assumed that IMME is correct. If one were to add a $c_3 T_Z^3$ term to Eq. (1), the systematic

TABLE III. Same as Table II but for the $T = 2$ quintet for $A = 32$. Experiment 1999 gives experimental values from [8].

Nucleus	T_Z	Experiment 1999	Experiment 2002	IMME 2002
^{32}Si	+2	−24 080.9(2.2)	−24 080.9(2.2)	−24 079.5(1.0)
^{32}P	+1	−19 232.88(20)	−19 232.88(20)	−19 232.92(20)
^{32}S	0	−13 970.98(41)	−13 970.98(41)	−13 970.80(38)
^{32}Cl	−1	−8296.9(1.2)	−8291.5(1.8)	−8293.14(68)
^{32}Ar	−2	−2180(50)	−2200.2(1.8)	−2200.0(1.6)
χ^2_ν	⋯	0.35	0.74	⋯
c_3	⋯	0.25(47)	−0.11(30)	⋯

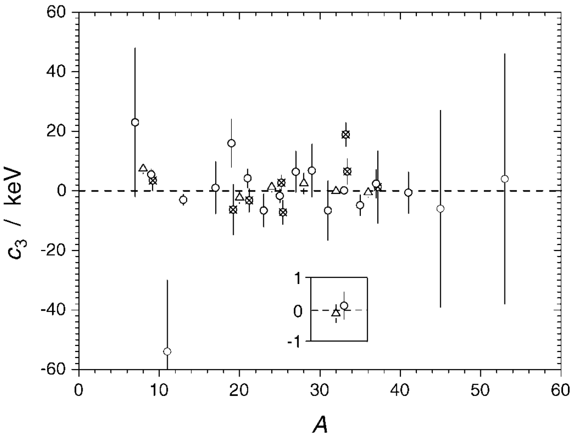

FIG. 2. The c_3 coefficient of all completely measured quartets (circles) and quintets (triangles) [3]. In the case of the quartets, the empty circles label the ground states; the crossed circles label the higher-lying states. The inset shows the values for $A = 32$ and $A = 33$ as obtained in this work. The c_3 scale is enlarged by a factor of 10.

uncertainty would grow to 0.0069 [8]. Since the result reported here implies a change of 9.1 keV with respect to the mass excess value $D(^{32}\mathrm{Ar}) = -2209.3$ keV used in [8], one can conclude that the correlation coefficient should be $a_{\mathrm{new}} = a_{\mathrm{old}} + \partial a/\partial Q \cdot \Delta Q = 0.9880$. However, the energy of the $T = 3/2$ resonance in ^{32}S, for which a significant disagreement with the previously accepted value was found [6], was used in [8] as a calibration for the ^{32}Ar spectrum. Recalibration of the ^{32}Ar spectrum affects three quantities that influence the correlation coefficient: (i) the value for the energy of the beta-delayed proton; (ii) the Q value for the beta-decay reaction; (iii) the energy calibration of the proton spectrum. The dependencies on (i) and (ii) are explicitly given in [8] to be $\partial a/\partial E_{\mathrm{cm}} = -0.9 \times 10^{-3}\ \mathrm{keV}^{-1}$ and $\partial a/\partial Q = -1.2 \times 10^{-3}\ \mathrm{keV}^{-1}$. The dependence on changing (iii) was omitted in [8] but was calculated [24] by one of the authors of [8]. The new experimental value for the decay energy is $Q = 6091.3(2.5)$ keV [which is in good agreement with the value 6093.1(1.4) obtained from the new IMME coefficients] and is 4.0 keV larger than the previous IMME value. Taking all this into account, one obtains

$$a = 1.0050 \pm 0.0052(\mathrm{stat}) \pm (\mathrm{syst}), \qquad (3)$$

where the statistical uncertainty from [8] was kept while the systematic uncertainty will be reduced since IMME is no longer used for the prediction of the mass of ^{32}Ar. A final value for the remaining systematic uncertainty can be given only after a reanalysis of the data with the changes mentioned above. Compared to the original work [8], these improved mass measurements and the IMME test reported in this work have put the measurement of the beta-neutrino correlation coefficient a on secure footing. The experimental determination of the mass of ^{32}Ar allows the measurement of the beta-neutrino correlation coefficient to stand by itself without having to rely on any mass model predictions.

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