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Breakdown of the Isobaric Multiplet Mass Equation at $A = 33$, $T = 3/2$

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Mass measurements on ^{33,34,42,43}Ar were performed using the Penning trap mass spectrometer ISOLTRAP and a newly constructed linear Paul trap. This arrangement allowed us, for the first time, to extend Penning trap mass measurements to nuclides with half-lives below one second (^{33}Ar: $T_{1/2} =$ 174 ms). A mass accuracy of about 10^{-7} ($\delta m \approx 4$ keV) was achieved for all investigated nuclides. The isobaric multiplet mass equation was checked for the $A = 33$, $T = 3/2$ quartet and found to be inconsistent with the generally accepted quadratic form.

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Since the strong or hadronic interaction is nearly charge independent, the isospin formalism is one of the basic tools in nuclear as well as particle physics. The neutron and the proton have isospin $T = 1/2$ with $T_Z^{\text{neutron}} = +1/2$ and $T_Z^{\text{proton}} = -1/2$. Every state of a nucleus has an isospin T and belongs to a $2T + 1$ multiplet formed by "analog" levels in different isobaric nuclei. The charge of each member is measured by its isospin projection $T_Z = (N - Z)/2$.

In light nuclei, isobaric analog states (IAS) have nearly identical wave functions. The charge-dependent energy difference of these states can be calculated in first-order perturbation theory assuming only two-body Coulomb forces. This leads to the simple equation, noted first by Wigner [1],

$$M(T_Z) = a + bT_Z + cT_Z^2, \qquad (1)$$

that gives the mass M of a member of an isospin multiplet as a function of T_Z. This quadratic relation is called the isobaric multiplet mass equation (IMME). It was thoroughly studied in the 1970s and reviewed by Benenson and Kashy in 1979 [2]. After looking at the quartets, it was found that IMME worked very well for 21 out of 22 cases. Because of its success and lack of experimental data, IMME is widely used to predict masses as well as level energies. For example, the IMME prediction of the mass of ^{32}Ar is essential for setting an upper limit on the scalar contribution to the weak interaction [3].

A more recent compilation of completely measured multiplets having $T \geq 3/2$, which serve to test the quadratic relationship given in Eq. (1), can be found in Ref. [4]. The lowest lying $A = 9$, $T = 3/2$ quartet is still the only significant exception for quartets. Here, a cubic term dT_Z^3 with $d = 5.5 \pm 1.7$ keV is required in order to describe the experimental data. Now, there are also six quintets with known masses. Only one of them, the $A = 8$, $T = 2$ quintet, does not agree with the quadratic form of IMME. Here at least one higher-order term has to be added to Eq. (1), either dT_Z^3, eT_Z^4, or both.

In this Letter, in addition to the masses of ^{34,42,43}Ar, we report on the direct measurement of the very short-lived nuclide ^{33}Ar ($T = 3/2$) that allows a further test of the IMME. The measurements were performed using the ISOLTRAP mass spectrometer [5] installed at the on-line mass separator facility ISOLDE/CERN [6]. The challenges of the measurements reported here were the very short half-life of 174 ms of ^{33}Ar and the limited production yield. Therefore, an efficient transfer of the ISOLDE ions into the trap as well as a fast measurement procedure had to be developed.

The argon isotopes were produced by bombarding a heated CaO target with bunches of 1.4 GeV protons delivered by the CERN proton-synchrotron-booster accelerator. The radionuclides produced were ionized in a plasma ion source, accelerated to 60 keV and mass separated by the ISOLDE general purpose mass separator. The integrated yield for ^{33}Ar was on the order of a few thousand ions per proton pulse, and for ^{34,42,43}Ar it was between 10 and 100 times higher.

For the ^{33}Ar measurement the ISOLDE beam gate was opened for a period of 30 ms about 70 ms after proton impact on the target. This scheme maximized the ^{33}Ar-to-background ratio. The ion beam was guided to the ISOLTRAP setup [5,7] shown in Fig. 1, which consists of three main parts: (i) a linear gas-filled radio frequency quadrupole (RFQ) trap [8] for retardation, accumulation,

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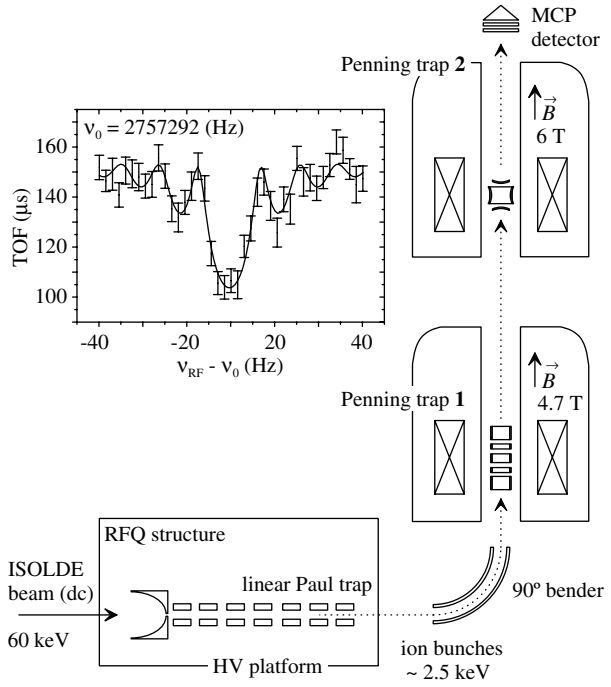

FIG. 1. Experimental setup of the ISOLTRAP mass spectrometer. The inset shows the cyclotron resonance curve for ^{33}Ar. The time of flight (TOF) of the ions from the trap to the ion detector is plotted as a function of the applied radio frequency ν_{rf}. The solid line is a fit of the theoretical line shape [10] to the data points.

cooling, and bunched ejection at low energy; (ii) a gas-filled cylindrical Penning trap for isobaric separation; (iii) a hyperboloidal Penning trap in ultrahigh vacuum for the actual mass measurement.

The 60-keV ISOLDE ion beam is first electrostatically retarded to an energy of a few eV and injected into the linear RFQ trap filled with helium buffer gas at about 10^{-2} mbar. The trap system consists of four segmented rods in a quadrupole arrangement to which rf and dc voltages are applied to achieve confinement of the ions. The ions entering the linear trap lose transverse and longitudinal energy due to collisions with the buffer gas. Within a cooling time of 2 ms the ions form a cloud in the potential well at the end of the system before they are extracted in a short pulse (FWHM $\approx 1\ \mu$s) by switching the potential of the last rod segments. The efficiency of the ISOLTRAP ion beam cooler and buncher was found to exceed 10%, in agreement with simulations, and a more than 10-fold reduction of the normalized ISOLDE beam emittance was achieved [8].

The low-energy Ar ion pulses are then captured in the first Penning trap. A mass selective buffer gas cooling technique [9] allows the operation of this trap as an isobar separator [7]. For the measurements presented here the ions spent 73 ms in the first Penning trap, where a mass resolving power of $R = 7000$ was achieved, sufficient to separate the Ar nuclei from their Cl and S isobars, also delivered by ISOLDE. After this cleaning procedure the Ar ions were transferred to the second Penning trap.

In the second trap the mass measurement is carried out via a determination of the cyclotron frequency $\nu_{\mathrm{c}} = \frac{q}{2\pi m}B$ of an ion with mass m and charge q in a magnetic field of strength B. The stable isotope ^{36}Ar having a well-known mass was used to calibrate the magnetic field at frequent intervals using the same measurement cycle as for the radioactive nuclides. The ion orbit radii were initialized for 10 ms before their cyclotron motion was excited for a period of $T_{\mathrm{rf}} = 60$ ms, yielding a linewidth of $\Delta\nu_{\mathrm{c}}(\mathrm{FWHM}) \approx 0.9/T_{\mathrm{rf}} = 15$ Hz. This results in a resolving power of $R = \nu_{\mathrm{c}}/\Delta\nu_{\mathrm{c}}(\mathrm{FWHM}) = 130\,000$.

In total, each cycle of the measurement of the cyclotron frequency took 175 ms. The overall efficiency of ISOLTRAP was about 10^{-4} given by the ratio of the number of ^{33}Ar ions detected after the second trap on a multichannel plate (MCP) detector to that in the ISOLDE beam. This efficiency includes decay losses, detection efficiency, and transfer losses. The main losses occur while injecting into the first Penning trap, where the ion optics has still to be improved.

The inset of Fig. 1 shows a cyclotron resonance curve for ^{33}Ar with a fit to the theoretical line shape [10]. Excellent agreement is observed. A total of about 2000 ions were detected by the MCP for this measurement, which took about eight hours, including the time required for reference measurements. The frequency ratio $\nu_{\mathrm{c}}(^{36}\mathrm{Ar})/\nu_{\mathrm{c}}(^{33}\mathrm{Ar})$ can be determined with a relative accuracy of 9×10^{-8}, governed by statistics and resolving power.

Table I summarizes the frequency ratios and their uncertainties for the investigated argon isotopes. The uncertainties are calculated by adding quadratically the statistical error and a conservative estimate of 1×10^{-7} for the relative systematic error that covers mainly unobserved magnetic field changes [11]. In the case of ^{33}Ar this results in a relative accuracy of 1.3×10^{-7} of the frequency ratio.

The frequency ratio $\nu_c^{\mathrm{ref}}/\nu_c$ is converted into an atomic mass value for the measured nuclide by

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

with the electron mass m_e and the mass of the reference nuclide m_{ref}. The resulting mass excesses are given in Table I together with literature values.

Our measurements improve the previous value of ^{33}Ar [12] by a factor of 7 in accuracy. Similar improvements are achieved for ^{42,43}Ar. The mass for ^{34}Ar agrees very well with the literature value demonstrating once more the reliability of ISOLTRAP measurements.

The $T = 3/2$ state quartets, to which ^{33}Ar belongs, are formed by the isobaric analog states in ^{33}S and in ^{33}Cl, ^{33}Ar, and ^{33}P. Until now the quadratic IMME consistency in this case was limited by the 30 keV uncertainty of the ^{33}Ar mass.

The mass of ^{33}P, measured by two groups from the β^- decay end point, was evaluated in [13]. The excited levels with $J^\pi = 3/2^+$ and $5/2^+$ were measured

TABLE I. Frequency ratios relative to ^{36}Ar and mass excesses (ME) for argon isotopes as determined in this paper and literature values from Ref. [13].

Nuclei	$T_{1/2}$	Frequency ratio ν_{ref}/ν	$\mathrm{ME}_{\mathrm{exp}}$[a] [keV]	$\mathrm{ME}_{\mathrm{lit}}$ [keV]
^{33}Ar	174 ms	0.917212520(126)	−9381.9(4.2)	−9380(30)
^{34}Ar	844 ms	0.944747261(105)	−18378.4(3.5)	−18378(3)
^{42}Ar	33 a	1.166694503(173)	−34422.7(5.8)	−34420(40)
^{43}Ar	5.37 m	1.194569791(159)	−32009.8(5.3)	−31980(70)

[a]Using ME(^{36}Ar) = −32454.927(29) μu [23] and 1 u = 931.494013 MeV/c^2 [24].

in ^{30}Si$(\alpha, p\gamma)^{33}$P and ^{31}P$(t, p\gamma)^{33}$P reactions consistently by three different groups [14].

The excitation energy for the $J^\pi = 1/2^+$ state in ^{33}S was measured in a number of reactions compiled in [14,15]. The most precise values result from the ^{32}S$(n, \gamma)^{33}$S reaction investigated by two groups giving $E_x = 5480.1 \pm 0.4$ keV [16] and $E_x = 5479.7 \pm 0.1$ keV [17]. The adopted value from [15] is 5480.1 ± 0.4 keV, but there is no reason given as to why the value from [17] is ruled out in favor of that from [16].

The $J^\pi = 1/2^+, 3/2^+, 5/2^+$ states in ^{33}Cl were studied in the ^{32}S$(p, p')^{32}$S reaction [18,19]. When recalculating the transformation of the proton energy from the laboratory frame to the center-of-mass frame, disagreement with the published values was found. The recalculation changed the mass excesses of these states by about 1 keV compared to the tabulated values [13,15], and the mass excess of the $J^\pi = 1/2^+$ state changes from −15459.5 ± 1.1 keV to −15460.1 ± 1.0 keV.

The excitation energies for the $J^\pi = 3/2^+$ and $5/2^+$ states in ^{33}S were measured consistently using ^{34}S$(p, d)^{33}$S and ^{32}S$(d, p)^{33}$S reactions [14].

Including the ISOLTRAP data, all members of the $A = 33$, $T = 3/2$ quartet are now very well known, allowing a stringent test of the quadratic IMME [Eq. (1)]. A least-squares fit of a quadratic function to the data in Table II results in $\chi^2 = 10.6$, meaning that the probability is only 0.1% that the data can be described this way. Allowing for an additional, cubic IMME term dT_Z^3 yields $d = -2.95 \pm 0.90$ keV (Table II) that is not consistent with zero.

Taking the excitation energy for the $J^\pi = 1/2^+$ state in ^{33}S from [17], instead of the adopted value from [15], results in $\chi^2 = 9.7$ or $d = -2.75 \pm 0.88$ keV. So, there is no significant change in the result due to this ambiguity.

By investigating the excited quartets for $A = 33$, one finds improved accuracy for the mass of the excited states of ^{33}Ar due to the improved accuracy for the ground state mass. Using the new ground state value, together with the excitation energies from [12], yields d coefficients of 18.9 ± 4.0 keV and 2.6 ± 4.0 keV for the $J^\pi = 3/2^+$ and $5/2^+$ quartets, respectively. The previously noted deviation from zero of the d coefficient of the $J^\pi = 3/2^+$ quartet [4] is now the most significant deviation from zero in all completely measured isospin quartets. However, it is very probable that the level assignment in ^{33}Cl is wrong for the $T = 3/2$, $J^\pi = 3/2^+$ state. The level width of this state is 2 orders of magnitude larger than the one for the other two levels and therefore is likely not the isobaric analog state with $T = 3/2$.

In Fig. 2 the d coefficients for all completely measured quartets are plotted together with the significance of their deviations from zero. All together there are five quartets with a d coefficient farther than two standard deviations away from zero and thus are in significant disagreement with the quadratic IMME, whereas statistics allows only 0.8 cases.

There has been great effort to explain a nonzero d coefficient, triggered by the $A = 9$ ground state quartet. The significant result for the $A = 9$ quartet has been partly explained by isospin mixing effects in the $T_Z = -1/2$ and $+1/2$ members [20,21], by the expansion of the least-bound proton orbit in ^{9}C, as well as by charge-dependent nuclear forces [22]. The difficulty lies in the fact that most of these second order effects are absorbed in the

TABLE II. Mass excesses of the levels of the $T = 3/2$ quartets for $A = 33$ and the results of the IMME test [χ^2 results from a fit of the quadratic IMME, as in Eq. (1), to the data, while d is the additional coefficient if IMME is assumed to be cubic].

Nucleus	T_Z	$\mathrm{ME}_{\mathrm{exp}}$ [keV] $J^\pi = 1/2^+$	$J^\pi = 3/2^+$	$J^\pi = 5/2^+$
^{33}P	+3/2	−26337.7(1.1)[a]	−24906.1(1.1)[b]	−24490.1(1.1)[b]
^{33}S	+1/2	−21106.14(41)[b]	−19681.2(3.0)[b]	−19249.2(4.0)[b]
^{33}Cl	−1/2	−15460.1(1.0)[c]	−14020.7(3.0)[c]	−13612.6(2.0)[b]
^{33}Ar	−3/2	−9381.9(4.2)[d]	−8038(20)[e]	−7596(20)[e]
χ^2	⋯	10.6	22.8	0.42
d	⋯	−2.95(90)	18.9(4.0)	2.6(4.0)

[a]From [13]. [b]From [13,15]; see also text. [c]From [13,18] but recalculating the center of mass energy of the protons; see text. [d]This work. [e]This work and [12].

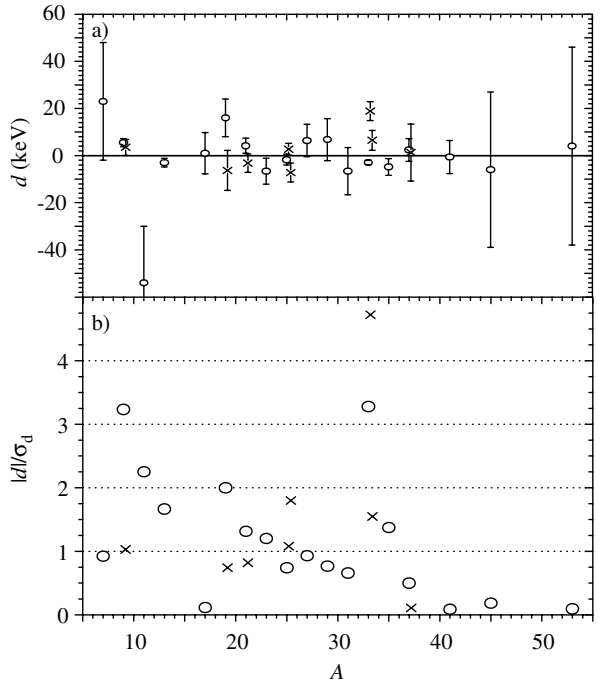

FIG. 2. (a) The d coefficients of all completely measured quartets and (b) the significance of the magnitude of their deviation from zero, $S = |d|/\sigma_d$; the circles label the ground state quartets, the crosses label the higher lying quartets. The data for the $A = 33$ quartets are from Table II; the other data are from Ref. [4].

b and c coefficients of Eq. (1) and therefore the theoretical approaches tend to produce a too small d coefficient. The discrepancy found in this paper for the $A = 33$ quartet imposes a new challenge to theory, requiring better calculations in the ^{33}Ar region. The stronger binding of ^{33}Ar and its higher Z enhance isospin mixing and charge-dependent nuclear force effects and should reduce the expansion of the least-bound proton orbit compared to ^{9}C.

In conclusion, even though it is not clear which effect causes the breakdown of IMME for the described $A = 33$, $T = 3/2$ quartets, it is necessary to be very careful if one derives high-accuracy masses of proton-rich configurations from the quadratic IMME. Such a case is, for example, the experiment searching for scalar weak interactions using the β decay of ^{32}Ar [3]. The result depends strongly on the mass of ^{32}Ar that was derived using IMME. If the $A = 32$ quintet required a d coefficient comparable to the one of the $A = 33$ quartet, the mass of ^{32}Ar would differ by about 20 keV from the value obtained with IMME and the result of the experiment would be overturned from agreement with the standard model to three-sigma evidence for scalar currents.

The presented measurements show for the first time that it is possible to measure the mass of charged particles with a half-life in the order of 100 ms using a Penning trap. At the same time the accuracy remains comparable to Penning trap mass measurements of longer-lived species. This was made possible by the development of a new efficient transfer of the ISOLDE ions into the trap using a RFQ ion beam cooler and buncher and a rapid measurement technique.

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