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

F. Herfurth[1], J. Dilling[1], A. Kellerbauer[2,5], G. Audi[4], D. Beck[6,1], G. Bollen[3,8], S. Henry[4], H.-J. Kluge[1], D. Lunney[4], R.B. Moore[5], C. Scheidenberger[1], S. Schwarz[2,8], G. Sikler[1], J. Szerypo[7] and the ISOLDE collaboration[2]

[1] GSI, Planckstraße 1, 64291 Darmstadt, Germany
[2] CERN, 1211 Geneva 23, Switzerland
[3] Sekt. Physik, Ludwig-Maximilians-Universität München, 85748 Garching, Germany
[4] CSNSM-IN2P3-CNRS, 91405 Orsay-Campus, France
[5] Department of Physics, McGill University, Montréal (Québec) H3A 2T8, Canada
[6] Instituut voor Kern- en Stralingsfysica, Celestijnenlaan 200 D, 3001 Leuven, Belgium
[7] Department of Physics, University of Jyväskylä, PB 35 (Y5), 40351 Jyväskylä, Finland
[8] NSCL, Michigan State University, East Lansing MI 48824-1321, USA
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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 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 (IMME) was checked for the $A = 33$, $T = 3/2$ quartet and found to be inconsistent with the generally accepted quadratic form.

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 $T_{Z}^{neutron} = +1/2$ and $T_{Z}^{proton} = -1/2$. Therefore, 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 $T_{Z} = (N - Z)/2$.

In light nuclei, levels with equal T, isobaric analog states have nearly identical wave functions. Therefore, the charge dependent energy difference of states with equal T in an isobaric series can be calculated in first-order perturbation theory under the assumption of 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}, the projection of the isospin T. This quadratic relation is called the isobaric multiplet mass equation (IMME). It was thoroughly studied in the 70's and reviewed by Benenson and Kashy in 1979 [2]. Looking at the quartets it was found that IMME worked very well for 21 out of 22 cases. The only exception was the most accurately known quartet, the $A = 9$, $T = 3/2$ quartet. Due to its success and due to the lack of experimental data, IMME is widely used to determine masses and level energies, especially of the members with the lowest T_{Z}.

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 reference [3]. 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.

To verify the predictions of IMME it is necessary to determine accurately the masses of all members of a multiplet with $T \geq 3/2$. For so called "ground state multiplets" the two states with the highest and lowest T_Z are the ground states of the concerned nuclides. These states are accessible to direct mass spectrometry.

In this letter we report on direct mass measurements on ^{33}Ar, which is a member of a $T = 3/2$ isospin quartet. Until now the test of quadratic IMME showed consistency for this quartet but was limited by the 30 keV uncertainty on the ^{33}Ar mass value. In addition, the masses of the isotopes ^{34,42,43}Ar were determined in this work. The measurements were performed by use of the ISOLTRAP mass spectrometer [4] installed at the on-line mass separator facility ISOLDE/CERN [5]. The particular difficulties of the measurements reported here were the very short half-life of 174 ms of ^{33}Ar and the limited production yield. Therefore, a fast measurement procedure as well as an efficient transfer of the ISOLDE ions into the trap 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 PS-Booster accelerator with a minimum distance of 1.2 s between two pulses. In the actual experiment between six and ten proton pulses per supercycle (total length 19.2 s) were sent to the ISOLDE target. The radionuclides produced were ionized in a plasma ion source and then mass separated by the ISOLDE GPS mass separator. The integrated yield for ^{33}Ar was in the order of a few thousand ions per proton pulse. The yield for ^{34,42,43}Ar was between 10 and 100 times higher.

For the ^{33}Ar measurement the ISOLDE beam gate was opened for a period of 30 ms with a delay of about 70 ms relative to the time of the proton impact on the target. This scheme maximized the ^{33}Ar-to-background ratio. The mass separated 60-keV ion beam was guided to the ISOLTRAP set-up shown in Fig. 1, which consists of three main parts: (1), a linear gas-filled radiofrequency quadrupole (RFQ) trap for retardation, accumulation, cooling and bunched ejection at low energy, (2), a gas-filled cylindrical Penning trap for isobaric separation, and (3), an ultra-high vacuum hyperboloidal Penning trap for isomeric separation and the actual mass measurement. The function and performance of the two Penning traps is described in [4,6] and the recently added linear Paul trap in [7].

The 60-keV ISOLDE argon 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 to which radiofrequency voltages are applied to obtain a transversely focusing force. The segmenta-

tion of the rods allows the creation of a DC electric field along the axis of the system. The whole system is built in UHV technology to suppress charge exchange reactions with gas impurities. 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 into a short ion pulse (FWHM $\approx$ 1 μs) by switching the potential of the last rod segments.

After having left the linear ion trap at a potential of 60 kV the Ar ion pulse is accelerated towards a cavity at a typical potential of 57.5 kV. When the ion pulse reaches the field-free region inside the cavity, the latter is switched to ground potential and the ions leave it with a kinetic energy of only 2.5 keV.

The efficiency of the ISOLTRAP ion beam cooler and buncher was found to exceed 10% in agreement with simulations. A more than 10-fold reduction of the normalized ISOLDE beam emittance has been achieved [7].

The low-energy Ar ion pulses are then electrostatically retarded and captured in the first Penning trap. The main task of this trap is the purification of the ion bunch from contaminating ions. A mass selective buffer gas cooling technique [8] is employed and allows the operation of this trap as an isobar separator [6]. For the measurements presented here the ions spent 73 ms in the first Penning trap. A mass resolving power of $R = 7000$ was achieved. This was sufficient to separate the Ar nuclei from their isobars Cl and S also delivered by ISOLDE. After this cleaning procedure, the Ar ions were pulsed out of the first Penning trap and transferred to the second one.

The second trap is a high-precision trap used for the actual mass measurement [4]. The measurement is carried out via a determination of the cyclotron frequency $\nu_c = \frac{q}{2\pi m} B$ of an ion with mass m and charge q in a magnetic field of strength B. The stable argon isotope ^{36}Ar having a well known mass was used to calibrate the magnetic field by measuring its cyclotron frequency. In the case of ^{33}Ar the ions were prepared for 10 ms before their cyclotron motion was excited for a time period of $T_{\mathrm{RF}} = 60$ ms yielding a line width of $\Delta\nu_c$(FWHM)$\approx 0.9/T_{\mathrm{RF}}$. This results in a resolving power of $R = \nu_c/\Delta\nu_c$(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 using a multi channel plate (MCP) detector, and those 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. On the average, less than one ^{33}Ar ion was stored in the precision trap in each cycle.

The inset of Fig. 1 shows a cyclotron resonance curve for ^{33}Ar with a fit to the theoretical line shape [9]. Ex-

cellent agreement is observed. About 2000 ions were detected by the MCP for this measurement, which took about eight hours including the time required to measure repeatedly the magnetic field using the reference isotope ^{36}Ar. The frequency ratio $\nu_c(^{36}\mathrm{Ar})/\nu_c(^{33}\mathrm{Ar})$ can be determined with a relative accuracy of $9 \cdot 10^{-8}$, governed by statistics and resolving power.

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

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

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

Our measurements improve the previous measurement of ^{33}Ar [11] by a factor of 7 in accuracy. Similar improvements are achieved for ^{42,43}Ar. The mass value for ^{34}Ar agrees very well with the literature value demonstrating once more the reliability of ISOLTRAP mass measurements.

The $T = 3/2$ state quartet, to which ^{33}Ar belongs, is formed by the isobaric analogue states in ^{33}S and ^{33}Cl, which have $J^{\pi} = 1/2^+$ and $T = 3/2$, and the ground states of ^{33}Ar and ^{33}P.

The mass of ^{33}P has been measured by two groups by determining the β^- decay endpoint and was evaluated in [12]. The excited levels with $J^{\pi} = 3/2+$ and $5/2+$ have been measured in $^{30}\mathrm{Si}(\alpha,\mathrm{p}\gamma)^{33}\mathrm{P}$ and $^{31}\mathrm{P}(\tau,\mathrm{p}\gamma)^{33}\mathrm{P}$ reactions consistently by three different groups [13].

The excitation energy for the $J^{\pi} = 1/2^+$ state in ^{33}S has been measured in a number of reactions compiled in [13,14]. The most precise values result from the $^{32}\mathrm{S}(\mathrm{n},\gamma)^{33}\mathrm{S}$ reaction investigated by two groups [15,16]. The excitation energies are $E_x = 5480.1(4)$ keV [15] and $E_x = 5479.7(1)$ keV [16]. The adopted value from [14] is 5480.1(4) keV, but there is no reason given as to why the value from [16] is ruled out in favor of that from [15].

The $J^{\pi} = 1/2^+$, $3/2^+$, $5/2^+$ states in ^{33}Cl have been measured by investigating the resonances in the $^{32}\mathrm{S}(\mathrm{p},\mathrm{p}')^{32}\mathrm{S}$ reaction [17,18]. Since this is the only reference for these states, both publications have been studied carefully. A difference was found in recalculating the transformation of the proton energy from the laboratory frame to the center of mass frame. The recalculation changed the mass excesses of these states by about 1 keV compared to the values found using [12,14]. For the $J^{\pi} = 1/2^+$ state the mass excess changed 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 $^{34}\mathrm{S}(\mathrm{p},\mathrm{d})^{33}\mathrm{S}$ and $^{32}\mathrm{S}(\mathrm{d},\mathrm{p})^{33}\mathrm{S}$ reactions [13].

Including the ISOLTRAP data all members of the $A = 33$, $T = 3/2$ quartet are very well known (see Tab. II). This allows a stringent test of the quadratic IMME, done by a least-square fit of Eq. 1 to the masses. The result is shown in Tab. II. The data are not very well described by the quadratic IMME relation: The fit of a quadratic function to the data results $\chi^2 = 10.6$ that has a probability of only 0.1% to occur if the data was described by a quadratic function. Allowing for an additional cubic term dT_Z^3 in IMME the data as given in Tab. II yields $d = -2.95 \pm 0.90$ keV, which is not consistent with zero.

If the excitation energy for the $J^\pi = 1/2^+$ state in ^{33}S is taken from ref. [16] instead of the adopted value in [14], the quadratic fit results $\chi^2 = 9.7$, while $d = -2.75 \pm 0.88$ keV.

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 [11] yields d-coefficients of 18.9 ± 4.0 keV and 6.5 ± 4.2 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 [3] is now the most significant deviation from zero in all completely measured isospin quartets.

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, 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 partly been explained by isospin mixing effects in the $T_Z = -1/2$ and $+1/2$ members [19,20], by the expansion of the least bound proton orbit in ^{9}C, as well as by charge-dependent nuclear forces [21]. However, the situation rests unclear.

In the case of the $A = 33$, $T = 3/2$ quartets, the Thomas-Ehrman shift has to be considered for the proton unbound states in ^{33}Cl. But, in Ref. [22] it has been shown, that this is practically fully absorbed in the a, b, and c-coefficients of Eq. 1 and that the contribution to a d-coefficient remains very small.

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. In the near future, ISOLTRAP will be used to measure the mass of ^{32}Ar which is an important input for $\beta - \nu$ correlation experiments [23]. For $A = 32$, belonging to the $T = 2$ quintet, an accurate experimental mass value of ^{32}Ar is still missing. A measurement by ISOLTRAP would improve the current uncertainty of 50 keV by at least a factor ten. This is a challenge due to the even shorter half-life and

lower production yield as compared to ^{33}Ar.

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TABLE I. Frequency ratios relative to ^{36}Ar and mass excesses (ME) for argon isotopes as determined in this work (with overall uncertainties) and literature values from Ref. [12].

nucl.	$T_{1/2}$	freq. ratio ν_{ref}/ν	ME$^*_{\mathrm{exp}}$ [keV]	ME$_{\mathrm{lit}}$ [keV]
^{33}Ar	174 ms	0.917212520(126)	-9381.9(42)	-9380(30)
^{34}Ar	844 ms	0.944747261(105)	-18378.4(35)	-18378(3)
^{42}Ar	33 a	1.166694503(173)	-34422.7(58)	-34420(40)
^{43}Ar	5.37 m	1.194569791(159)	-32009.8(53)	-31980(70)

*using ME(^{36}Ar) $= -32454.927 \pm 0.029\,\mu$u [24] and $1\,$u $= 931.494013\,$MeV/c^2 [25].

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 is 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).

nucl.	T_{Z}	ME$_{\mathrm{exp.}}$ [keV]		
		$J^\pi = 1/2^+$	$J^\pi = 3/2^+$	$J^\pi = 5/2^+$
^{33}P	+3/2	-26337.7(11)[a]	-24906.1(11)[b]	-24490.1(11)[b]
^{33}S	+1/2	-21106.14(41)[b]	-19681.2(30)[b]	-19249.2(40)[b]
^{33}Cl	−1/2	-15460.1(10)[c]	-14020.7(30)[c]	-13604.8(30)[c]
^{33}Ar	−3/2	-9381.9(42)[d]	-8038(20)[e]	-7596(20)[e]
χ^2		10.6	22.8	2.4
d		-2.95(90)	18.9(40)	6.5(42)

[a]from [12]; [b]from [12,14], see also text; [c]from [12,17] but recalculating the center of mass energy of the protons, see text; [d]this work; [e]this work and [11]
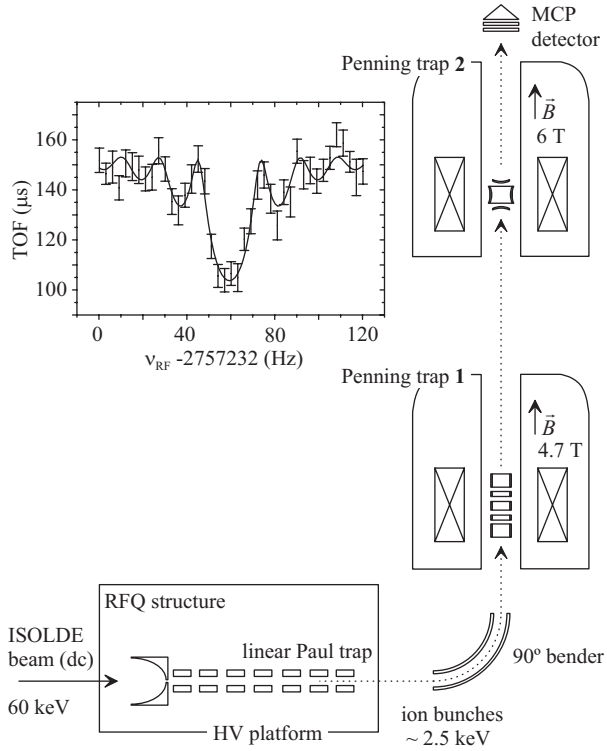
FIG. 1. Experimental setup of the ISOLTRAP mass spectrometer. The inset shows the cyclotron resonance curve for ^{33}Ar. Plotted is the time of flight (TOF) of the ions from the trap to the ion detector as a function of the applied radio frequency. The solid line is a fit of the theoretical line shape [9] to the data points.
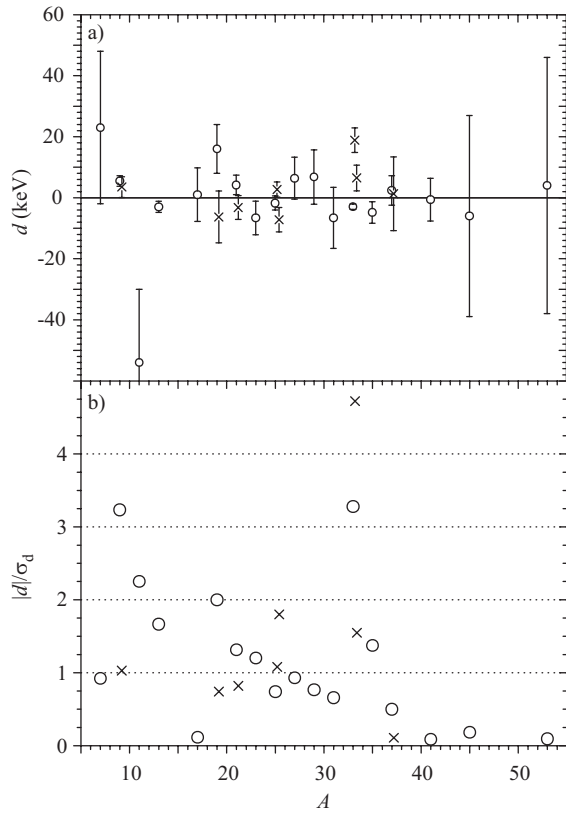
FIG. 2. The d-coefficients of all completely measured quartets (a) and the significance of their deviation from zero $S = |d|/\sigma_\mathrm{d}$ (b); the circles label the ground state quartets, the crosses the higher lying quartets. The data for the $A = 33$ quartets are from Tab. II, the other data are from Ref. [3].