Mass Measurements of the Neutron-Deficient ^{41}Ti, ^{45}Cr, ^{49}Fe, and ^{53}Ni Nuclides: First Test of the Isobaric Multiplet Mass Equation in fp-Shell Nuclei

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Mass measurements of the neutron-deficient ^{41}Ti, ^{45}Cr, ^{49}Fe and ^{53}Ni nuclides: First test of the isobaric multiplet mass equation in *fp*-shell nuclei

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Isochronous mass spectrometry has been applied to neutron-deficient ^{58}Ni projectile fragments at the HIRFL-CSR facility in Lanzhou, China. Masses of a series of short-lived $T_z = -3/2$ nuclides including ^{41}Ti, ^{45}Cr, ^{49}Fe, and ^{53}Ni have been measured with a precision of $20-40$ keV. The new data enable for the first time to test the isobaric multiplet mass equation (IMME) in fp-shell nuclei. We observe that the IMME is inconsistent with the generally accepted quadratic form for the $A = 53$, $T = 3/2$ quartet. We perform full space shell model calculations and compare them with the new experimental results.

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Isospin symmetry, a fundamental concept in nuclear and particle physics, allows to classify states with quantum numbers T and T_z in addition to, e.g., spin J and parity π. Within the isospin formalism, protons (p) and neutrons (n) are described as different charge states of the nucleon with total isospin value $T = 1/2$, but having the isospin projections of $T_z(p) = -1/2$ and $T_z(n) = +1/2$, respectively. All states in a nucleus composed of Z protons and N neutrons have the same isospin projection $T_z = (N-Z)/2$ but they can have different total isospin $T = |N-Z|/2, |N-Z|/2+1, \ldots$. States in isobaric nuclei with the same T and J^π that have very similar structure and properties can be considered as members of an isobaric multiplet. These isobaric analog states (IAS) are energetically degenerate in the absence of any charge-dependent nucleon-nucleon interaction when the neutron-proton mass difference is corrected for. In general, there are several isobaric multiplets for a set of isobaric nuclei.

Assuming the two-body nature for any charge-dependent effects and the Coulomb force between the nucleons, Wigner [1] as well as Weinberg and Treiman [2] noted that masses, m, of the $2T+1$ members of an isobaric multiplet are related by the isobaric multiplet mass equation (IMME):

$$\mathrm{ME}(A,T,T_z) = a(A,T) + b(A,T)T_z + c(A,T)T_z^2, \quad (1)$$

where ME$= (m - A\cdot u)c^2$ is the mass excess value and a, b, c are parameters depending on the atomic mass number A and the total isospin T. Extra terms such as dT_z^3 or eT_z^4 can be added to IMME in order to provide a measure of any deviation from the quadratic form associated with isospin symmetry. Numerous measurements have been performed investigating the validity of IMME. Reviews and compilations of existing data can be found in Refs. [3, 4].

In the recent years, precision tests of IMME became possible due to access to accurate mass data coming mainly from Penning trap facilities. The tests were focused on the light-mass region [5–15]. As a general trend, quadratic form stems well with the data [3, 4], except for slight disagreements at $A = 8$, 9, 32, and 33 [3–6, 12–16]. Several explanations for the cubic term in IMME have been suggested such as isospin mixing, second-order Coulomb effects, or charge-dependent nuclear forces [5, 16–18]. To our knowledge, no experimental tests of IMME have been reported in *fp*-shell up to now. The main reason for this is obviously the lack of accurate mass data on exotic $T_z = -3/2, -1/2$ nuclei. As noted in Ref. [17], the correction to the quadratic form of IMME with the introduction of $d(A,T)T_z^3$ in Eq. (1) is proportional to $Z\alpha$ (α being the fine structure constant). Hence, the effects of isospin mixing and/or charge-dependent nuclear forces may be enhanced in heavy nuclei. We like to stress that an accurate test of IMME in *fp*-shell is motivated not only by the fundamental importance of isospin symmetry, but also by the requirements of accurate mass predictions for the neutron-deficient nuclei in this region, which in turn is essential, e.g., for understanding the astrophysical rp-process of nucleosynthesis [19].

In this Letter, we report on the new mass measurements conducted at the Institute of Modern Physics in Lanzhou, China. Masses of a series of $T_z = -3/2$ nuclei have been determined with high accuracy. In particular, the masses of ^{41}Ti, ^{45}Cr, ^{49}Fe and ^{53}Ni nuclides enable us to perform the first experimental test of IMME in the fp-shell. We observe that for

the $A = 53$ ($T = 3/2$) quartet IMME is inconsistent at a 3.5σ confidence level with the generally accepted quadratic form.

The experiment was conducted at the HIRFL-CSR accelerator complex. Its high-energy part consists of the synchrotron CSRm, the fragment separator RIBLL2, and the cooler-storage ring CSRe [20]. To produce short-lived $T_z = -3/2$ nuclei of interest, we used projectile fragmentation of 463.36 MeV/u ^{58}Ni primary beams in a $\sim$15 mm ^{9}Be production target. At this energy, the reaction products emerge from the target as bare nuclei, i.e. with no atomic electrons. After in-flight separation with RIBLL2, the cocktail beam of exotic nuclei within a Bρ-acceptance of about $\pm$0.2% was injected into CSRe. Both RIBLL2 and CSRe were set to a fixed magnetic rigidity of $B\rho = 5.6770$ Tm to allow for an optimal transmission of the $T_z = -3/2$ nuclides centered around ^{47}Mn. Other nuclides within the acceptance of the RIBLL2-CSRe system were transmitted and stored as well. Typically, about ten ions were stored simultaneously from each injection.

The masses of stored ions were measured employing the Isochronous Mass Spectrometry (IMS) technique [21–23]. In this technique, the ring is tuned into the isochronous ion-optical mode such that the velocity spread of injected ions is compensated by their orbit lengths. As a result, the revolution times of the ion becomes a direct measure of its mass-over-charge ratio, m/q (see Refs. [21–23] for details).

To measure revolution times of stored ions, we used a timing detector [24], which is equipped with a 19 μg/cm^2 carbon foil of 40 mm in diameter installed inside CSRe aperture. Secondary electrons were released from the foil at each passage of every stored ions. The electrons were guided to a set of micro-channel plates (MCP), thus giving timing signals. The latter were directly sampled using a digital oscilloscope. For each injection the recording time was set to 200 μs, which corresponding to $\approx$320 revolutions of the ions. The periodic timing signals were used to determine the revolution time of each ion. The revolution times of all ions form a revolution-time spectrum. More details can be found in Ref. [23, 25].

The resolving power of CSRe mass spectrometry is determined by the instabilities of magnetic fields which cause small shifts of the entire revolution time spectra measured for different injections (see Ref. [25]). Compared to our previous measurements [23, 25], the stability of the magnets has been improved significantly. Furthermore, the magnetic fields of CSRe dipole magnets were constantly monitored which was used to identify time intervals of relatively constant magnetic fields. Different from the data analysis described in Ref. [25], the entire data, accumulated in a 5-days experiment, were grouped according to these time intervals. In total 761 independent sub-spectra were obtained. Each spectrum corresponds to about 100 injections into CSRe. Taking the relative shifts between individual spectra into account, the 761 sub-spectra were combined into a common revolution time spectrum. Figure 1 illustrates a part of this spectrum zoomed in a time window of 608 ns $\leq t \leq$ 620 ns. The identification of the peaks in the spectrum was done in the same way as in Refs. [25, 26]. The standard deviations of the revolution-time

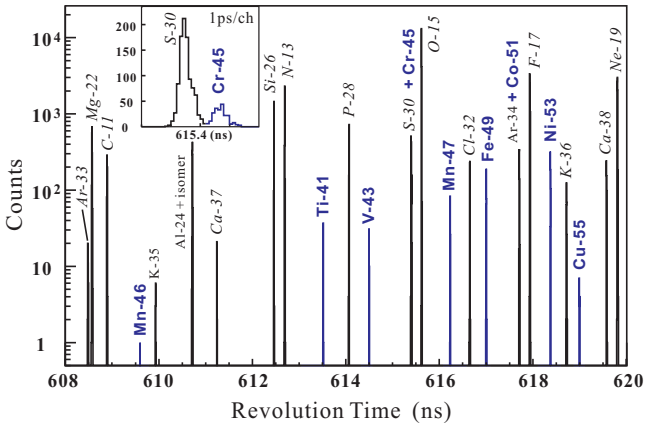

FIG. 1: The revolution time spectrum zoomed in the time window of 608 ns $\leq t \leq$ 620 ns. The insert shows the well-resolved peaks of ^{30}S^{16+} and ^{45}Cr^{24+} nuclei, which have very similar m/q values. Nuclei with masses determined in this experiment and those used as references are indicated with bold and italic letters, respectively.

TABLE I: Experimental ME values obtained in this work and values from the updated atomic-mass evaluation AME'11 [27]. The extrapolated values are indicated with symbol "#". The deviations $\delta = \mathrm{ME_{CSRe}} - \mathrm{ME_{AME'11}}$ are given in the last column. Also listed are the numbers of identified ions N, standard deviations σ_t and FWHM values of the revolution time peaks (see Fig. 1). The latter are converted in keV via FWHM$= 2.36 \cdot q \cdot (a_1 + 2a_2 \cdot t + 3a_3 \cdot t^2) \cdot \sigma_t$, where a_1, a_2 and a_3 are the free parameters of the calibration fit.

Atom	N	σ_t (ps)	FWHM (keV)	$\mathrm{ME_{CSRe}}$ (keV)	$\mathrm{ME_{AME'11}}$ (keV)	δ (keV)
^{41}Ti	76	2.0	580	$-15698(28)$	$-15090(363)$	$-608(364)$
^{45}Cr*	218	2.2	702	$-19515(35)$	$-19403(196)^{\#}$	$-112(199)$
^{49}Fe	338	3.0	1026	$-24751(24)$	$-24824(149)^{\#}$	73(151)
^{53}Ni	651	4.1	1488	$-29631(25)$	$-29687(298)^{\#}$	56(299)

* see text and Ref. [31].

peaks in this time range lie within $2 \sim 5$ ps and the achieved mass resolving power amounts to $m/\Delta m = 180000$. In order to calibrate the spectrum, fourteen nuclides (see Fig. 1) with accurately known masses [27] were used to fit their m/q values versus the revolution time t by employing a third order polynomial. The unknown mass values were determined by interpolating the fit function to the corresponding times t.

In order to estimate possible systematic errors, we re-determined the ME values of each of the fourteen reference nuclides by calibrating the spectrum with the other thirteen nuclides. The agreement between our re-determined ME values and the corresponding literature ones has been examined by calculating the normalized χ-value, defined as $\chi_n = \sqrt{\chi^2/n}$ with $n = 14$ in our case, in the same way as in our previous measurements [23, 25]. The obtained $\chi_n = 1.18$ is within the expected range of $\chi_n = 1 \pm 0.19$ at 1σ confidence level, indicating that no additional systematical errors have to be considered. The ME values of ^{41}Ti, ^{45}Cr, ^{49}Fe, and ^{53}Ni determined in this work are listed in Table I.

A low-lying isomeric state in ^{45}Cr ($E_x = 107$ keV, $T_{1/2} >$

TABLE II: Compilation of ME values for ground states (g.s.), isobaric analog states (IAS) and the corresponding excitation energies (E_x) for $A = 41, 45, 49$, and 53 ($T = 3/2$) quartets. Also listed are χ_n for quadratic fits and d-coefficients for cubic fits (see text).

Atom	T_z	ME(g.s) (keV)	E_x (keV)	ME(IAS) (keV)
^{53}Ni	$-3/2$	$-29631(25)^{**}$	0	$-29631(25)$
^{53}Co	$-1/2$	$-42658.6(17)$ [27]	4393(19) [33]	$-38266(19)$ [33]
^{53}Fe	$+1/2$	$-50946.7(17)$ [27]	4250(3) [32]	$-46696.7(34)$
^{53}Mn	$+3/2$	$-54689.0(6)$ [27]	0	$-54689.0(6)$
Quadratic fit: $\chi_n = 3.7$				
Cubic fit: $d = 39(11)$				
^{49}Fe	$-3/2$	$-24751(24)^{**}$	0	$-24751(24)$
^{49}Mn	$-1/2$	$-37615(24)$ [27]	4809(28) [33]	$-32806(15)$ [33]
^{49}Cr	$+1/2$	$-45333(2)$ [27]	4764(5) [32]	$-40569(5)$
^{49}V	$+3/2$	$-47961.0(9)$ [27]	0	$-47961.0(9)$
Quadratic fit: $\chi_n = 1.5$				
Cubic fit: $d = 13.2(89)$				
^{45}Cr	$-3/2$	$-19515(35)^{**}$	0	$-19515(35)$
^{45}V	$-1/2$	$-31880(17)$ [27]	4791(19) [33]	$-27089(9)$ [33]
^{45}Ti	$+1/2$	$-39008.3(8)$ [27]	4723(7) [32]	$-34285(7)$
^{45}Sc	$+3/2$	$-41070.4(6)$ [27]	0	$-41070.4(6)$
Quadratic fit: $\chi_n = 0.7$				
Cubic fit: $d = 5.4(82)$				
^{41}Ti	$-3/2$	$-15698(28)^{**}$	0	$-15698(28)$
^{41}Sc	$-1/2$	$-28642.41(8)$[27]	5937(3) [33]	$-22705(3)$ [33]
^{41}Ca	$+1/2$	$-35137.92(14)$ [27]	5819(2) [32]	$-29320(2)$
^{41}K	$+3/2$	$-35559.544(4)$ [27]	0	$-35559.544(4)$
Quadratic fit: $\chi_n = 0.6$				
Cubic fit: $d = -2.8(50)$				

** this work.

80 μs) has been reported in Ref. [30]. This isomer can not be resolved in our spectra. Therefore, a dedicated analysis was conducted to account for a possible contamination by the isomer. Details of the analysis and the error propagation will be reported elsewhere [31]. The outcome is to increase the uncertainty from 20 to 35 keV.

Our ME(^{41}Ti)$= -15698(28)$ keV is in excellent agreement with ME(^{41}Ti)$= -15700(100)$ keV recommended in the atomic-mass evaluation AME'03 [28]. However, it differs largely from ME(^{41}Ti)$= -15090(360)$ keV measured in the storage ring ESR of GSI using the same IMS technique [29]. Compared to Ref. [29], we accumulated by a factor of 15 larger counting statistics for ^{41}Ti, achieved by a factor of 1.6 higher mass resolving power, and, very essentially, we have used 14 well-known reference masses instead of 4.

To test the validity of the quadratic form of IMME, the energies of four members of a $T = 3/2$ multiplet are required. These are the mass values of the ground states of $T_z = \pm 3/2$ nuclei and the IAS energies of the $T_z = \pm 1/2$ nuclei. Note, that the spin and parity of the $T = 3/2$ IASs for $A = 45, 49$, and 53 are $J^\pi = 7/2^-$ and for $A = 41$ they are $J^\pi = 3/2^+$.

With our new mass values, the data of four $T = 3/2$ isospin quartets at $A = 41, 45, 49$, and 53 are completed for the first time. All available experimental data for these multiplets are compiled in Table II. The mass values for the ground states are from this work and from Ref. [27]. The IAS excitation energies in the $T_z = +1/2$ nuclei are from compilation [32]. The data for the $T_z = -1/2$ nuclei were obtained in Ref. [33] from measurements of β-delayed protons of the respective $T_z = -3/2$ nuclei. Please note, those listed in Table II, ME(IAS) and E_x values from Ref. [33], are updated here taking into account the most recent ground-state masses compiled in Ref. [27].

Assuming the quadratic form of IMME (see Eq. (1)), the fit results for $A = 41, 45$, and 49 have reasonable χ_n values (see Table II). A striking result $\chi_n = 3.7$ is obtained for the $A = 53$, $T = 3/2$ isobaric multiplet. This corresponds to a probability of 0.02% that the data can be described by Eq. (1). Using very accurate data for ^{53}Fe and ^{53}Mn, we have recalculated the ME(IAS) values for ^{53}Ni and ^{53}Co. Since three values are needed to fit a parabolic function, we used in addition the mass of ^{53}Ni to determine the value for ^{53}Co, and vice versa. The corresponding results are ME(^{53}Ni)$= -29397(58)$ keV and ME(^{53}Co)$= -38344(9)$ keV. Both values deviate by 4σ from the values in Table II. Therefore, we added a cubic term dT_z^3 in Eq. (1) and derived all four coefficients from the four ME values. In particular, the b and d coefficients for the $T = 3/2$ states are given by the differences $b = (9b_{3,3} - b_{3,1})/8$ and $d = (b_{3,1} - b_{3,3})/2$, where $b_{2T,2T_z} = [\mathrm{ME}(A,T,-T_z) - \mathrm{ME}(A,T,T_z)]/(2T_z)$. When $b_{3,3} = b_{3,1}$ we have $d = 0$ and $b = b_{3,3} = b_{3,1}$.

The obtained d coefficients are given in Table II and presented in Figure 2 together with recent precision tests in the sd-shell nuclei [9–12]. For the $A = 53$ ($T = 3/2$) quartet we obtain $d = 39(11)$ keV which deviates by 3.5σ from zero thus indicating a dramatic failure of the quadratic form of IMME.

We note that no long-lived states are known in ^{53}Mn [35]. Thus, assuming mirror symmetry which works well in nuclear structure, no long-lived isomers are expected in ^{53}Ni and the measured ME value should correspond to the ground state. In the unlikely case that an unknown isomer would exist in ^{53}Ni, the ground-state ME value would inevitably be more negative than the reported one, thus leading to a more pronounced breakdown of IMME, i.e., to an even larger d coefficient.

Experimental and theoretical $b_{3,1}$, $b_{3,3}$ and d coefficients for the case of $A = 53$ are given in Table III. The theory is based upon two isospin non-conserving (INC) Hamiltonians. The simplest one is that from the $f_{7/2}$ model space considered in [34], where the $(f_{7/2})^2$ two-body matrix elements for the proton-proton, neutron-neutron and proton-neutron interactions were obtained from a fit to the isobaric displacement energies in nuclei with $A = 41 \sim 55$. The rms deviation between experiment and theory was 12 keV. The results given in Table III are from Ref. [34]. The difference $d = (b_{3,1} - b_{3,3})/2$ can only come from isospin mixing, and in the $f_{7/2}$ model it is less than 1 keV, in disagreement with $d = 39(11)$ keV.

The second result is obtained for the full ($f_{7/2}$, $f_{5/2}$, $p_{3/2}$, $p_{1/2}$) (pf) model space. The Hamiltonian is composed of the GPFX1A isospin conserving Hamiltonian [36–38] plus the Ormand-Brown (OB) isospin non-conserving Hamiltonian [39]. Details of the GPFX1A part and its applications to many

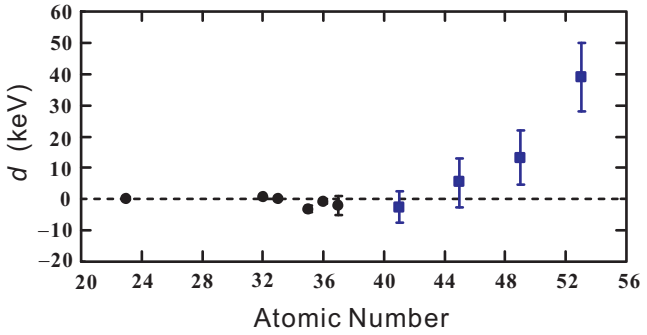

FIG. 2: d coefficients for the four $T = 3/2$ isobaric multiplets in pf-shell (squares). Experimental data since 2001 (circles) [9–12] are shown for comparison. Please note, that albeit with large uncertainties, there seems to be a trend of gradual increase of d with A in fp shell.

TABLE III: $b_{3,1}$, $b_{3,3}$ and d coefficients for $A = 53$.

	b_{33} (keV)	b_{31} (keV)	d (keV)
Experiment	8353(8)	8431(19)	39(11)
Theory ($f_{7/2}$ model)	8366	8365	−0.5
Theory (full pf model)	8292	8290	−1.0

pf shell data are given in Refs. [36–38]. The OB part was obtained from a consideration of Coulomb, charge-symmetry breaking and charge-dependence breaking interactions with strengths determined from the b and c coefficients for nuclei with $A = 41 \sim 59$. The theoretical value $d(pf) = -1.0$ keV does not agree with experiment. The numerical error in the d coefficient is less than one keV, while the main theoretical error comes from the position of the $T = 1/2$ states. There are several nearby $T = 1/2$ states that could mix with the $T = 3/2$ state with a level density of about one per 150 keV. But the typical isospin mixing matrix element is on the order of 5 keV or less, so it is impossible to get an energy shift of the $T = 3/2$ state due to two-level mixing of more than about 5 keV.

Inspection of the b values in Table III shows that $b_{3,1}(\mathrm{exp}) = 8431(19)$ keV is higher than other values. The largest uncertainty in this value comes from the determination of the excitation energy of the $T = 3/2$ state in ^{53}Co obtained from the energy of β^+-delayed protons of ^{53}Ni [33]. To change the $d(\mathrm{exp})$ value from 39 keV to zero, the proton energy would require to be lowered by 78 keV from 1929 keV to 1851 keV. A new proton-decay experiment with a higher resolution is highly desirable to address the issue.

In summary, we have accurately measured masses of a series of $Tz = -3/2$ nuclei from ^{41}Ti through ^{53}Ni. This allowed us for the first time to perform a test of IMME in fp-shell nuclei. We found a breakdown of the quadratic form of IMME for the $A = 53$ ($T = 3/2$) quartet. The disagreement cannot be explained by neither the existing nor the new theoretical calculations of isospin mixing. If this breakdown can be confirmed by improved experimental data (ground-state masses, energies of the IAS), possible reasons, such as enhanced effects of isospin mixing and/or charge-dependent nuclear forces in the fp-shell, should be investigated.

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