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Precision test of the isobaric multiplet mass equation for the $A = 32$, $T = 2$ quintet

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Masses of the radionuclides ^{32,33}Si and ^{34}P and of the stable nuclides ^{32}S and ^{31}P have been measured with the Low Energy Beam and Ion Trap (LEBIT) Penning trap mass spectrometer. Relative mass uncertainties as low as 3×10^{-9} have been achieved. The measured mass value of ^{32}Si differs from the literature value by four standard deviations. The precise mass determination of ^{32}Si and ^{32}S have been employed to test the validity of the quadratic form of the isobaric multiplet mass equation (IMME) for the most well known $A = 32$, $T = 2$ isospin quintet. The new experimental results indicate a dramatic breakdown of the model.

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More than seventy years have passed since Heisenberg first proposed the concept of isospin [1]. Introduced to describe explicitly the charge independence of the nuclear force, it is treated like a quantum number used in nuclear and particle physics. A quarter century later, Weinberg and Treiman noted that the masses of an isospin multiplet should lie along a parabola [2]:

$$M(T_z) = a + bT_z + cT_z^2, \tag{1}$$

where $T_z = (N - Z)/2$. The isobaric multiplet mass equation (IMME), as it is now known, is based on the assumption that the charge-dependent differences in nucleon mass arise from pair interactions. Higher order terms, dT_z^3 and eT_z^4, require second-order Coulomb effects, charge-dependent nucleon-nucleon interaction, or many-body forces. Numerous studies of isospin multiplets have confirmed the validity of the quadratic form of IMME [3], and significant deviations have only occurred in light multiplets with unbound states (such as $A = 9, T = 3/2$). In addition to the fundamental importance of isospin symmetry, another feature of IMME is the prediction of unmeasured (or insufficiently precise) masses for astrophysical applications, e.g., mapping the proton drip line for the rp-process [4], or tests of physics beyond the standard model, e.g., searching for scalar currents [5]. The most precise test of IMME was made with the $A = 32$, $T = 2$ quintet [3], but recent measurements [6–8] first upheld and then invalidated the quadratic form of IMME. The most recent discussion [8] contained speculation on isospin mixing in ^{32}S or on an erroneous ^{32}Si mass. An analysis of the data indicates that if ^{32}Si were more bound by 3 keV, then the quadratic form would be restored.

In this Rapid Communication, we report a direct mass measurement of radioactive ^{32}Si by Penning trap mass spectrometry. The technique has already been used to test the validity of the quadratic form IMME in several cases [7,9–11], including measurements at the Low Energy Beam and Ion Trap (LEBIT) facility at the National Superconducting Cyclotron Laboratory (NSCL) [9].

In the present work, the radioactive isotopes ^{32,33}Si and ^{34}P were produced by fragmentation of a 140 MeV/nucleon primary ^{40}Ar beam at the NSCL Coupled Cyclotron Facility. The A1900 fragment separator [12] was used to separate secondary 94.5 MeV/nucleon fragments that were transported to the LEBIT facility. The radionuclides were slowed in solids and then thermalized in a linear gas cell [13] by collisions with helium atoms at a pressure of 600 mbar. A series of ring electrodes within the gas cell were used to guide ions to the extraction nozzle where gas flow ejected them into a differentially pumped radio-frequency quadrupole (RFQ) ion guide system [14]. The last section of the ion guide was operated as a mass filter to select ions by their mass number to charge state ratio, A/Q; hence, only the ions of interest and molecular isobars exited the ion guides. Various residual gas impurities can be ionized by the beam or form a molecular ion with the stopped isotope [9,14]. For example, the isobaric contaminant $O_2^+(A/Q = 32)$ had a much higher rate than ^{32}Si$^+$ ions at the Penning trap. The simultaneous storage of several ions in the Penning trap can lead to systematic errors in a mass determination and must be suppressed [15]. Activity was measured after the mass filter with ^{33}Si, which was found to come as bare, singly

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charged ions. Therefore, water vapor was introduced into the gas cell to create radiomolecular ions with $A/Q = 33, 68, 86$, consistent with $^{33}Si^+$ and $^{33}SiOH(H_2O)_n^+$, $n = 1, 2$. For the best yield, $n = 2$ was selected. Similar chemistry, $^{34}P(H_2O)_2^+$, was observed for ^{34}P ions that were part of the $^{33}Si/^{34}P$ cocktail beam delivered by the A1900 fragment separator.

The other mass measurements reported in this Rapid Communication are of stable ions, $^{32}S^+$ and $^{31}P^+$. Both masses had been measured with Penning trap mass spectrometry at Florida State University with relative uncertainties below 10^{-9} [16,17]. Therefore, our measurement of these isotopes served as an accuracy check of our method. Note that ^{32}S is the $T_z = 0$ member of the $A = 32$, $T = 2$ quintet. It is found as an impurity in the helium gas and ionized by an RF discharge in the gas cell. Sulfur formed HSO_2^+ in the presence of and SO_2^+ in the absence of water vapor. ^{31}P ions were produced in a plasma ion source from heated $^{31}P_2O_5$ powder from a small reservoir; $^{31}P^+$, P_2^+, and HPO_2^+ were extracted from this source.

After exiting either the ion guides or the test ion source, the beam next entered a helium-gas-filled RFQ cooler and buncher [18]. There, a water molecule was stripped from the $SiOH(H_2O)_2^+$ and $^{34}P(H_2O)_2^+$ molecular ions, resulting in $SiOH(H_2O)^+$ and $^{34}P(H_2O)^+$. All ^{32}S and ^{31}P molecular ions emerged intact from the cooler and did not undergo collision-induced dissociation with the buffer gas.

The beam cooler and buncher produced ion pulses approximately 100 ns long, which were sent to the LEBIT Penning trap [19] housed in a $B = 9.4$ T superconducting solenoid. A fast kicker operated as a time-of-flight mass separator, and a dynamic capture process further purified the beam to a single mass unit based on the time-of-flight of ions from the cooler and buncher to the Penning trap. Any remaining isobaric contaminants were removed in the Penning trap by exciting their reduced cyclotron motion before measurement of the true cyclotron frequency of the ion of interest. To further minimize any mass shifts, measurements were performed with a low number of stored ions, averaging less than one ion at a time, with the exception of the $^{31}P^+$ and O_2^+ measurements, whose average number of ions were 5 and 6, respectively. The time-of-flight cyclotron resonance detection technique [20] was used to measure the cyclotron frequency $\nu_c = B \cdot q/2\pi m$, from which the ion mass m was determined. A typical resonance curve from this work is shown in Fig. 1.

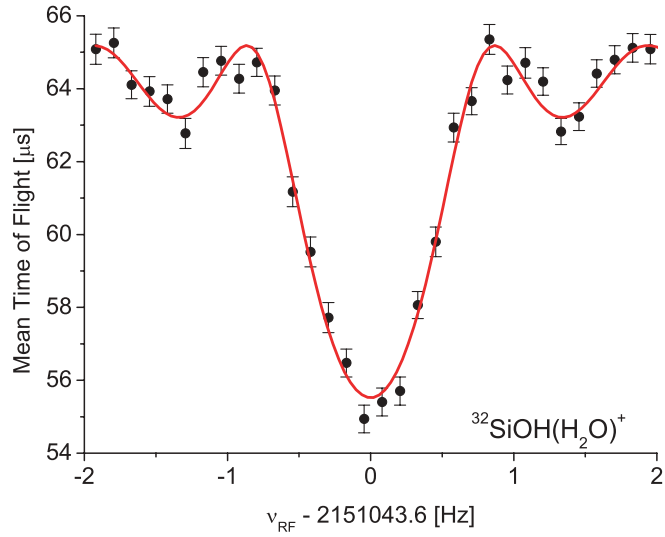

FIG. 1. (Color online) Mean time-of-flight as a function of the applied RF frequency in the Penning trap, ν_{RF}. The theoretical line shape [21] fitted to the data is shown by the solid curve.

Measurements of the ion of interest were interleaved with those of a reference ion to calibrate the magnetic field. The primary experimental result is thus the frequency ratio $R^i = \nu_c^i/(\nu_{ref}^{int})^i$, where $(\nu_{ref}^{int})^i$ is linearly interpolated from the reference measurements that bracket the measurement of ν_c^i. Table I lists the weighted average values of $\bar{R}$ for all ions. In the case of the ^{32}S, which was measured with reference ions CF_3^+ and $H_2CO_2F^+$, the mass was first extracted from the individual ratios, and then a weighted average was taken.

The reference ions were chosen with A/Q as close to that of the ion of interest as possible to minimize any mass-dependent systematic effects. The effect on the ratio R has been found to be less than $5(5) \times 10^{-10}$/u for the LEBIT system [9]. The drift of the magnetic field was monitored, and nonlinear temporal field changes were negligible compared to the statistical error [9]. Relativistic effects due to ion motion in the trap cancel if the ion of interest and its reference have the same A/Q and experience the same electric and magnetic forces. The largest relativistic shift was calculated for $^{31}P^+$ ($\Delta A/Q = 1$ relative to its reference O_2^+), which affected the ratio on the order of 2×10^{-10}, again negligible compared to the

TABLE I. For each mass measured, the isotope is listed with its molecular ion, its reference ion, the number of measurements N, the mean frequency ratio $\bar{R}$, and the relative uncertainty.

Isotope	Molecular ion	Reference ion	N	$\bar{R}$	$\delta\bar{R}/\bar{R}$
^{32}Si	$^{32}SiOH(H_2O)^+$	$C_5H_7^+$	13	1.001 004 962 8(48)	4.8×10^{-9}
^{33}Si	$^{33}SiOH(H_2O)^+$	$^{13}C^{12}C_4H_7^+$	7	1.000 983 204(11)	1.1×10^{-8}
^{31}P	$^{31}P^+$	O_2^+	3	1.032 804 708 2(32)	3.1×10^{-9}
^{34}P	$^{34}P(H_2O)^+$	$C_4H_5^+$	5	1.020 293 195(17)	1.7×10^{-8}
^{32}S	$^{32}SO_2H^+$	CF_3^+	8	1.061 959 895 0(55)	5.2×10^{-9}
	$^{32}SO_2H^+$	$H_2CO_2F^+$	7	1.000 525 733 9(88)	8.8×10^{-9}
	$^{32}SO_2^+$	CF_3^+	3	1.078 692 991(10)	9.3×10^{-9}

TABLE II. Measured mass excesses and a comparison to literature values.

Isotope	ME_{LEBIT} (keV)	ME_{lit} (keV)	ΔME[a] (keV)
^{32}Si	−24077.68(30)	−24080.86(77)[b]	3.18(82)
^{33}Si	−20514.30(70)	−20493(16)[c]	−22(16)
^{32}S	−26015.34(32)	−26015.535(2)[d]	0.20(32)
^{31}P	−24440.53(09)	−24440.541(1)[e]	0.01(9)
^{34}P	−24548.71(81)	−24558(5)[b]	9(5)

[a] $\Delta ME = ME_{LEBIT} - ME_{lit}$.
[b] Avogadro project, PTB [22].
[c] AME'03 [23].
[d] Penning trap measurement, FSU [16].
[e] Penning trap measurement, FSU [17].

statistical uncertainty. To account for unidentified impurities, we analyzed data for ion-number-dependent frequency shifts [15]. Only in the cases of $^{33}SiOH(H_2O)^+$, $^{32}SO_2H^+$, $^{32}SO_2^+$, and $^{31}P^+$ were significant frequency shifts observed. The maximum ratio shift was 2.2×10^{-9} (for the $^{32}SO_2^+/CF_3$ ratio), and corresponding corrections were applied to the frequency ratio. See Table I for the final ratios. The uncertainties listed in Tables I and II include statistical and all applicable systematic errors.

Table II contains a comparison of our results with values found in either the 2003 Atomic Mass Evaluation (AME'03) [23] or more recent experimental data. For the masses of stable ^{32}S and ^{31}P, we find excellent agreement with recent Penning trap mass measurements [16,17] performed with a single ion technique that achieved a precision of $\lesssim$1 eV. All of the present mass measurements of unstable nuclei agree with the literature values except ^{32}Si. The AME'03 mass value of −24080.81(05) keV was previously determined by a series of (n,γ) reactions starting with ^{28}Si [24]. The authors themselves later published another value with a larger uncertainty of −24080.86(77) [22] without clarification. We observe a 4σ deviation from the value in Ref. [22] and find that ^{32}Si is 3 keV less bound than formerly thought. Our measurement is, however, in agreement with an older (t, p) study only published as a conference abstract [25]. In the case of ^{33}Si, previous values had been determined by transfer reactions, e.g., ^{34}S(^{14}C,^{17}O) [26] and ^{36}S(^{11}B,^{14}N) [27], and by β-γ coincidence techniques [28]. Both techniques result in uncertainties in the tens of keV, while the present uncertainty is more than 20 times smaller. In the case of ^{34}P, a (d, α) reaction was used to determine the mass, although the authors of AME'03 increased its uncertainty from 1.2 keV to 5 keV; we achieve a factor of 6 smaller uncertainty than the AME'03 value and a slightly smaller value.

To test the quadratic form of IMME, we fitted a parabola to the values of the ^{32}Si mass and the best available data for the four other members of the $A = 32$, $T = 2$ quintet (in decreasing T_z): $ME(^{32}Si) = -24077.68(30)$ keV, $ME(^{32}P) = -19232.78(20)$ keV, $ME(^{32}S) = -13967.58(28)$ keV, $ME(^{32}Cl) = -8283.47(6.61)$ keV, and $ME(^{32}Ar) = -2200.2(1.8)$ keV. Ground state masses were taken from Refs. [7,16,23] and this work, while the energies of the excited $T = 2$ states are from Refs. [8,29,30]. The difference

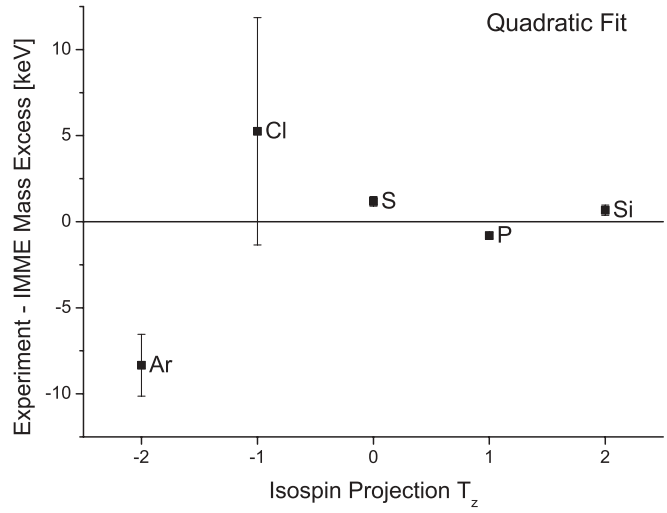

FIG. 2. Residuals of a quadratic IMME fit ($\chi^2/n = 31$) for the $A = 32$, $T = 2$ quintet in keV. Ground state masses are taken from Refs. [16,23] except for ^{32}Si, whose mass is from this work. Excited $T = 2$ energy levels are from Refs. [8,29,30].

of each value from the fitted function is shown in Fig. 2. With $\chi^2/n = 31$, we find a significant deviation from the quadratic form of IMME, caused by the observed 3 keV reduction in the binding energy of ^{32}Si. This shift is opposite in direction to that required to restore the quadratic form. A significant cubic $d = 1.00(9)$ keV term is needed to bring the reduced chi-square with this data set to an acceptable level ($\chi^2/n = 0.48$), as Fig. 3 illustrates.

A nonzero cubic (or quartic) coefficient in IMME could indicate isospin mixing of the $T = 2$ excited state in ^{32}Cl, ^{32}S, or ^{32}P with a nearby state. In general, d coefficients arising from isospin mixing have been theoretically predicted to reach values up to ≈1 keV [31], but no detailed calculation for the $A = 32$, $T = 2$ quintet has been reported yet.

Meaningful interpretation of breakdowns of the quadratic form of IMME require the availability of accurate and precise

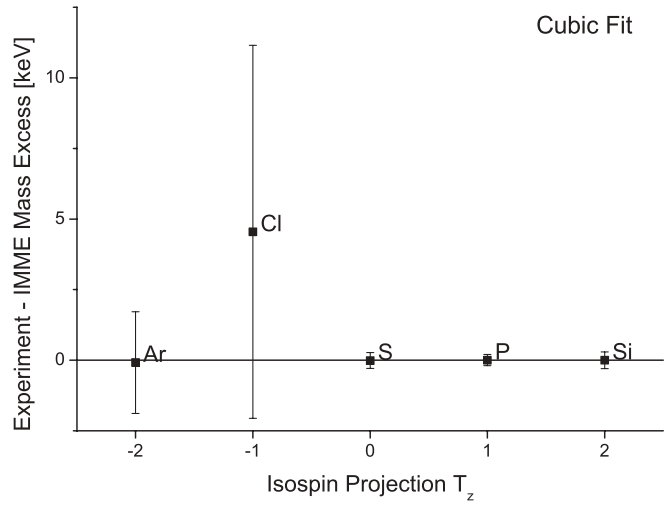

FIG. 3. Same as Fig. 2, but for a cubic IMME fit ($\chi^2/n = 0.48$).

masses for the members of the multiplet. In the $A = 32$, $T = 2$ quintet, the ground states of three members (^{32}Si, ^{32}S, ^{32}Ar) have been measured directly with high-precision Penning trap mass spectrometry. The new measurements of ^{31}P and ^{32}S confirm not only the precision but also the accuracy of this technique. The ground state masses of ^{32}P or ^{32}Cl have not been measured directly. The former has been determined precisely with neutron capture and β-decay measurements, all in agreement [23]. The energy of the first $T = 2$ level is also well known from neutron capture studies [32]. Unfortunately, data on ^{32}Cl do not agree with each other. The AME'03 [23] ground state mass value is based on charge exchange data. A β-delayed proton energy measurement [5] is being reevaluated [30,33] and differs from the AME'03 value by $\approx$7 keV. We have chosen to use the conservative AME'03 value for our fit. For the energy of the first $T = 2$ excited state, we use the most recent value [30]. Although in disagreement with older data [29], the new value's precision is an order of magnitude better. (The most precise values for the ground state masses of ^{31}P and ^{32}S from Refs. [16,17] change the literature values by less than 150 eV.) We should also note that the measurements of the excited $T = 2$ state energy of ^{32}S are somewhat inconsistent (see Ref. [8] for discussion).

In summary, we performed the first direct measurements of the masses of ^{33}Si and ^{34}P, reducing their mass uncertainty to less than 1 keV. Our measurement of the mass of ^{32}Si with an accuracy of less than 0.5 keV provides a new test of the validity of the quadratic form of IMME. We find an even more dramatic breakdown of the quadratic form of IMME than the authors of Ref. [8], which may be explained by isospin mixing; a more likely cause, given the observed inconsistencies, is an erroneous mass value for ^{32}Cl or, less likely, a wrong value for an excited state energy in a non-ground-state multiplet member. Clearly, improved experimental tests of IMME in this and other multiplets are required to obtain reliable and precise information on isospin symmetry breaking in nuclei. Penning trap mass measurements do and will continue to play a decisive role.

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