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# High-precision mass measurement of $^{31}$S with the double Penning trap JYFLTRAP improves the mass value for $^{32}$Cl

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The mass of $^{31}$S has been measured with the JYFLTRAP double Penning trap mass spectrometer. The new mass excess value of $-19\,042.55(24)$ keV deviates from the adopted value of $-19\,044.6(15)$ keV by $1.4\sigma$. The mass value of $^{32}$Cl has been revised with the new $^{31}$S result and the latest data from $\beta$-delayed proton decay of $^{32}$Cl. The new mass excess value for $^{32}$Cl is $-13\,334.88(65)$ keV, which is the most precise value for $^{32}$Cl so far and in agreement with the recent ($^{3}$He,$t$) data. The isobaric multiplet mass equation has been tested in the $T=2$ quintet at $A=32$, and the cubic form of the equation has been found to agree with the experimental data. The $Q_{EC}$ value for the $\beta$ decay of $T=1/2$ mirror nucleus $^{31}$S has been determined as $Q_{EC}=5397.99(24)$ keV, which slightly deviates from the previously adopted value.

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*Introduction.* $^{31}$S ($Z=16, N=15$) is a $T_Z=-1/2$ mirror nucleus. Its mass excess (ME), $\beta$ decay $Q_{EC}$ value, and proton separation energy $S_p$ are essential for several reasons. The adopted ME value of $^{31}$S $-19\,044.6(15)$ keV in the Atomic Mass Evaluation 2003 (AME03) [1] is solely based on a $^{32}$S($p,d$)$^{31}$S $Q$ value [2]. The mass of $^{31}$S has been used for an accurate determination of the $^{32}$Cl mass value from the $\beta$-delayed proton data [3]. A recent $^{32}$S($^{3}$He,$t$)$^{32}$Cl measurement [4] showed a deviation from the mass value of $^{32}$Cl based on the $^{32}$Cl proton separation energy [3] and the adopted mass of $^{31}$S [1]. A possible reason for this could be an incorrect $^{31}$S mass value in Ref. [1]. As the mass of $^{32}$Cl is crucial for testing the isobaric multiplet mass equation (IMME) in the isospin $T=2$ quintet at mass $A=32$, it is important to study this discrepancy via a direct mass measurement of $^{31}$S.

$^{31}$S $\beta$ decays to its mirror nucleus $^{31}$P. Recently, corrected $ft$ values for these mirror transitions have been calculated [5,6]. Corrected $ft$ values can be used for a determination of Gamow-Teller to Fermi mixing ratios $\rho$ [5]. If the mixing ratio is already known, for example, via the determination of the $\beta$-neutrino angular correlation $a_{\beta\nu}$, such as for $^{21}$Na [7], the $|V_{ud}|$ value for the Cabibbo-Kobayashi-Maskawa matrix can be extracted from the corrected $ft$ value [6]. For an accurate determination of the corrected $ft$ values, the $Q_{EC}$ value has to be known with high precision. In addition to the $^{31}$S $Q_{EC}$ value, an improved mass value of $^{32}$Cl, and, thus, a more precise $Q_{EC}$ value for $^{32}$Ar, is needed in the studies of positron-neutrino correlations in the superallowed $0^+ \to 0^+$ $\beta$ decay of $^{32}$Ar [8].

The proton separation energy of $^{31}$S is important in the modeling of explosive hydrogen burning in ONe novae. The calculated reaction rate for a resonant proton capture on $^{30}$P depends exponentially on the proton separation energy of $^{31}$S. Therefore, even a small change in the proton separation energy will have an effect on the rate. The reaction $^{30}$P($p,\gamma$)$^{31}$S plays a major role governing the flow toward $^{32}$S and heavier species in nova nucleosynthesis [9,10]. The effect of the results of this Rapid Communication on the resonant capture rate will be discussed in Ref. [11].

*Experimental method.* The $^{31}$S ions were produced at the Ion-Guide Isotope Separator On-Line (IGISOL) [12] facility via $^{32}$S($p,pn$)$^{31}$S reactions with a 40-MeV proton beam, which impinges on a thin 2-mg/cm$^2$ ZnS target. The ions, which typically have a charge state $q=+e$ were accelerated to 30 keV, mass separated, and injected into the radio-frequency quadrupole [13], which cools the ions and delivers them as short bunches to JYFLTRAP [14], the double Penning trap mass spectrometer at IGISOL. In the first trap of JYFLTRAP, the purification trap, isobaric purification via mass-selective buffer-gas cooling [15] is performed. In the second trap, the precision trap, the mass is determined via the time-of-flight (TOF) ion-cyclotron resonance method [16,17] (see Fig. 1). The cyclotron frequency $\nu_c$ of an ion with a charge $q$, depends on the magnetic field $B$ and the mass of the ion $m_{\mathrm{ion}}$: $\nu_c = qB/(2\pi m_{\mathrm{ion}})$. By measuring the frequency ratio between a well-known reference ion and the ion of interest, the mass of the nuclide of interest can be obtained: $m=(\nu_c^{\mathrm{ref}}/\nu_c^{\mathrm{ion}})\times(m_{\mathrm{ref}}-m_e)+m_e$. Here, the obvious choice for the reference ion of $^{31}$S is its $\beta$-decay daughter $^{31}$P, which has been measured with very high precision at Florida State University, $m(^{31}\mathrm{P}) = 30.973\,761\,9989(9)$ u [18], and is simultaneously produced at IGISOL.

The mass of the ion of interest is obtained via scanning the sideband frequency $\nu_+ + \nu_-$, which corresponds to the cyclotron frequency at very high precision [19]. Since the motion is, at first, purely magnetron, it will maximally be converted to the cyclotron motion when the excitation frequency matches the sideband frequency. The ions in resonance gain more radial energy and experience a stronger axial force in the magnetic-field gradient when extracted from the trap, and, thus, will arrive at the microchannel plate detector faster than when off-resonance. With the Ramsey method of separated oscillatory fields [20,21], high precision for the measured

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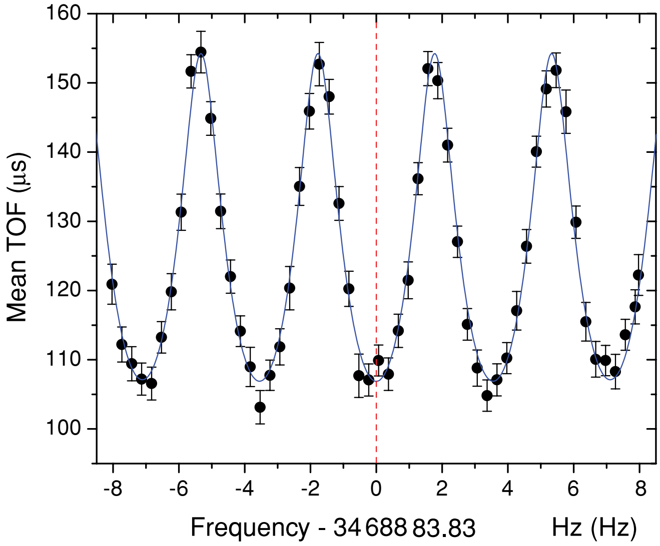


FIG. 1. (Color online) A TOF spectrum of $^{31}$S measured at JYFLTRAP. Only the ions that correspond to the first class (one to three ions) of the first 30-min interval of the first $^{31}$S file are shown. The total number of ions in the figure is 2002.

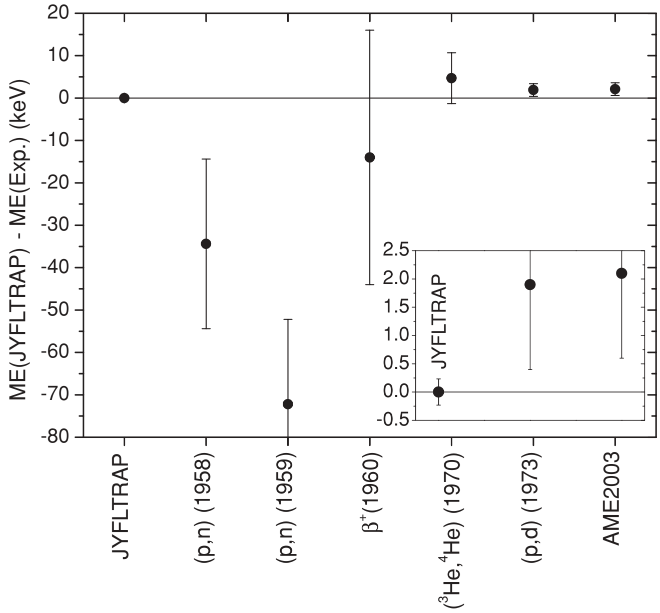


FIG. 2. A comparison of the new JYFLTRAP ME value of $^{31}$S to the previous measurements based on $(p,n)$ threshold energies [29,30], $\beta^+$ end-point energy [26], $^{32}$S($^3$He,$^4$He)$^{31}$S [27], $^{32}$S$(p,d)^{31}$S [2], and the AME03 value [1]. The inset shows the JYFLTRAP, $^{32}$S$(p,d)^{31}$S [2], and the AME03 values. The most recent mass values of $^{31}$P [18] and $^{32}$S [28] have been used in the calculations.

cyclotron frequencies was achieved by employing a timing pattern 25–250 ms (wait) – 25 ms.

The measured frequencies have been corrected for the count-rate effect [22]. For the fluctuations in the magnetic field, a correction of $\delta_B(\nu_{\mathrm{ref}})/\nu_{\mathrm{ref}} = 5.7(8) \times 10^{-11}$ min$^{-1}$ $\Delta t$, where $\Delta t$ is the time between the two reference measurements, has been quadratically added to the statistical uncertainty of each frequency ratio. Here, interleaved scanning [23] to minimize the effects of temporal fluctuations has been applied. The data have been split into about 30-min intervals, and the resonance frequencies have been fitted separately for $^{31}$S and $^{31}$P for each interval to obtain the frequency ratio. The weighted mean of the measured 23 frequency ratios has been calculated. The inner and outer errors [24] of the data set have been compared, and the larger value, in this case, the inner error, has been taken as the error of the mean. An additional residual relative error of $\delta_{\mathrm{res,lim}}(r)/r = 7.9 \times 10^{-9}$ from detailed carbon cluster measurements performed at JYFLTRAP [25] has been quadratically added to the data.

*Results and discussion.* Mass excess values of $^{31}$S and $^{32}$Cl—Altogether 23 measured frequency ratios were obtained, which included 107 585 ions of $^{31}$S and 99 032 ions of $^{31}$P in total. The measured frequency ratio is $r = 1.000\,187\,096\,6(15)(80)$, which shows the error without and with the additional relative residual uncertainty [25]. The obtained value for the ME of $^{31}$S is $-19\,042.55(24)$ keV, which deviates by 2.1(15) keV from the adopted value $-19\,044.6(15)$ keV.

The new mass value agrees with the old $\beta^+$ end-point measurement [26] and a $Q$-value measurement of $^{32}$S($^3$He,$^4$He)$^{31}$S [27], which were neglected in the AME2003 analysis. The adopted value [1] is based on the $Q$ value of $^{32}$S$(p,d)^{31}$S [2]. Both the AME03 value and the $(p,d)$ value with the new $^{32}$S mass [28] disagree with the JYFLTRAP value (see Fig. 2). The old $(p,n)$ threshold energy measurements [29,30] are clearly off from the present-day values and have not been included in the AME03 either.

With a proton separation energy of $^{32}$Cl $S_p(^{32}\mathrm{Cl}) = 1581.3(6)$ keV [3] and the new ME value for $^{31}$S, a revised ME value of $-13\,334.88(65)$ keV is obtained for $^{32}$Cl. The new value agrees with the adopted AME03 value [1] and with the earlier data based on $(p,n)$ threshold energy [31] and $^{32}$S($^3$He,$t$)$^{32}$Cl measurements [4,32] but disagrees with an old $(p,n)$ threshold energy measurement [33]. The result also shows that the discrepancy between the recent ($^3$He,$t$) data [4] and the data based on the proton separation energy of $^{32}$Cl and the ME of $^{31}$S from the AME03 [1] can be explained by the error in the ME value of $^{31}$S [1] (see Fig. 3).

IMME for the $T = 2$ multiplet at $A = 32$—According to the IMME, the members of an isobaric multiplet should lie along a parabola,

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

where $T_Z = (N - Z)/2$. The IMME is based on the assumption that isospin is a good quantum number, and the members of an isobaric multiplet should have equal energies in the absence of Coulomb interaction. The breakdown of the quadratic form of IMME [see Eq. (1)] at $A = 32$ was found in Ref. [34] and was confirmed in Ref. [4]. A possible error in the ME value of $^{32}$Cl or in the $T = 2$ excitation energies has been proposed to be responsible for the breakdown of the IMME at $A = 32$ [34]. In general, suggested explanations for a cubic term in the IMME have been isospin mixing, second-order Coulomb effects, or charge-dependent nuclear forces [35,36].

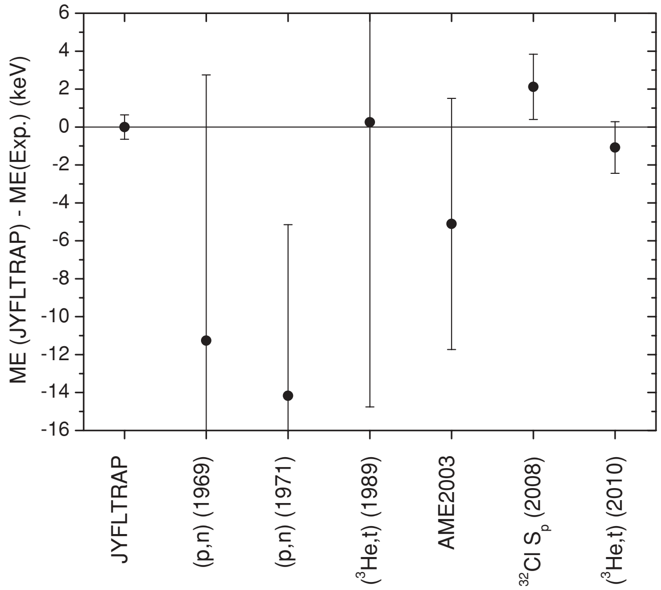


FIG. 3. The ME value of $^{32}$Cl compared to the previous measurements based on $(p,n)$ threshold energies [31,33], $^{32}$S($^3$He, $t$)$^{32}$Cl [32], AME2003 [1], $S_p$ of $^{32}$Cl [3], and the $^{31}$S mass from Ref. [1], and a recent $^{32}$S($^3$He,$t$)$^{32}$Cl measurement [4]. The value of this Rapid Communication was determined via the measured mass of $^{31}$S and the proton separation energy $S_p$ of $^{32}$Cl [3]. The most recent mass value of $^{32}$S [28] has been used in the calculations.

To test the IMME with the new $^{32}$Cl ME value, a parabola was fitted to the best available ME data of the $T = 2$ quintet at $A = 32$ (see Table I). The best available data for $^{32}$S, $^{32}$Cl, and $^{32}$Ar are consistent, but the data for $^{32}$Si and $^{32}$P are controversial. There is a huge deviation of 3.2(3) keV(10.6$\sigma$) in the ME of $^{32}$Si, which depends on whether it is taken from the $^{28}$Si ground-state mass [18] and the precisely measured neutron separation energies ($S_n$) of $^{29-32}$Si [1], or from the mass measurement performed at the Low Energy Beam and Ion Trap (LEBIT) [34]. The mass value given in the Avogadro project [37] is also based on $(n,\gamma)$ values of the silicon isotopes, but there, a much larger error is given without any further comments on possible systematic error. It is difficult to find a reason for this discrepancy. The Penning trap measurements [18,34] should be precise. On the other hand, the $S_n$ values of $^{29-31}$Si are based on many consistent precise $(n,\gamma)$ measurements performed (e.g., at McMaster [43,44], Los Alamos [45], and Institut Laue-Langevin [46]). Only the value for $^{32}$Si is based on a single measurement [46].

TABLE I. The ME values for the ground states and the excitation energies of the $T = 2$ states at $A = 32$. The data sets that correspond to the different $^{32}$Si and $^{32}$P values are labeled (A–F).

| Nuclide | $T_Z$ | Set | ME$_{gs}$ (keV) | $E_x(T = 2)$ (keV) |
|---|---|---|---|---|
| $^{32}$Si | 2 | A,B | −24 080.92(5)[a] | 0 |
| | | C,D | −24 077.68(30) [34] | 0 |
| | | E,F | −24 080.86(77) [37] | 0 |
| $^{32}$P | 1 | A,C,E | −24 304.94(12)[b,c] | 5072.48(9)[c] [38,39] |
| | | B,D,F | −24 305.22(19) [1] | 5072.44(6) [40] |
| $^{32}$S | 0 | | −26 015.5346(15) [28] | 12 047.96(28) [41] |
| $^{32}$Cl | −1 | | −13 334.64(57)[d] | 5046.3(4) [3] |
| $^{32}$Ar | −2 | | −2200.2(18) [42] | 0 |

[a]ME value of $^{28}$Si [18] and $S_n$ values for $^{29}$Si–$^{32}$Si [1] used.
[b]ME value of $^{31}$P [18] and $S_n$ value for $^{32}$P [38,39] used.
[c]An additional systematic error of 20 ppm was taken into account also for Ref. [38] where only a statistical error was given.
[d]A weighted mean of this Rapid Communication and Ref. [4].

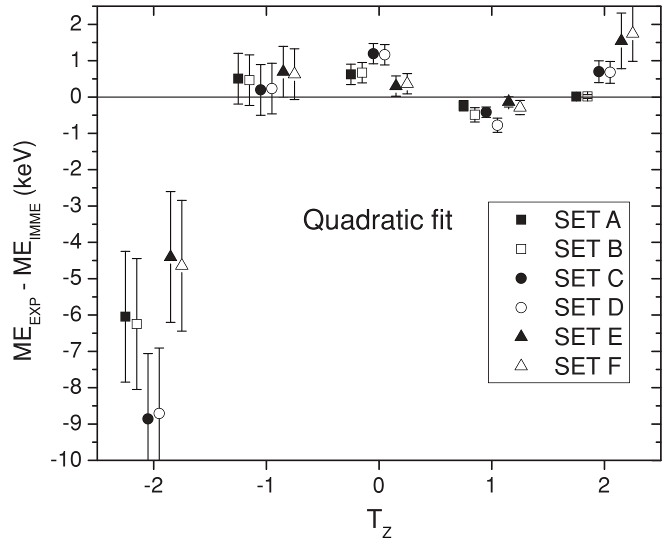


FIG. 4. Differences for the error-weighted quadratic fits of the mass excess values in the $T = 2$ quintet at $A = 32$. The error bars represent only the uncertainties of the experimental mass excess values given in Table I. The corresponding data sets are given in Table I, and obtained $\chi^2/n$ values are given in Table II.

A quadratic fit was performed with six different data sets, which correspond to the three different values for $^{32}$Si and two values for $^{32}$P (see Fig. 4). The fits with two different values for $^{32}$P (full and open symbols) do not differ much, but the fits with different $^{32}$Si values vary a lot. However, the error-weighted quadratic fit fails in all data sets. The smallest $\chi^2/n$ value is obtained with the $^{32}$Si value from the Avogadro project [37] and the $^{32}$P from Refs. [18,38,39] (data set E). The biggest deviations to the fit are seen at $^{32}$Ar in all data sets. If the $^{32}$Ar value was about 3$\sigma$(5.4 keV) higher, the quadratic fit with the data set E would yield a $\chi^2/n = 1.7$. The remeasurements of argon isotopes at ISOLTRAP have changed the mass excess values for $^{33}$Ar [42,47] by −2.2 keV and for $^{34}$Ar [47,48] by 1.3 keV, respectively. To confirm the breakdown of the IMME, a new mass measurement of $^{32}$Ar would be desirable.

The LEBIT mass excess value for $^{32}$Si [34] yields the highest $\chi^2/n$ values for quadratic fits but surprisingly low $\chi^2/n$ values for the cubic fits (see Fig. 5). In fact, the fit with the LEBIT $^{32}$Si [34] value and the $^{32}$P value from Refs. [18,38,39] (data set C) gives a $\chi^2/n = 0.002$, and the deviation from the fit is less than 0.016 keV for all nuclides. As can be seen from Table II, the cubic coefficients are very sensitive to the values used for $^{32}$Si and $^{32}$P mass excesses. Obviously, more direct mass measurements for the $T = 2$ quintet at $A = 32$ are needed for final verification of the breakdown of the IMME and the value of the cubic coefficient.

TABLE II. Obtained $\chi^2/n$ values for the quadratic and cubic IMME fits of the $T = 2$ quintet at $A = 32$. The value for the cubic coefficient $d$ is also tabulated.

| Set | $\chi^2/n_{\text{quadr.}}$ | $\chi^2/n_{\text{cubic}}$ | $d$ (keV) |
|---|---|---|---|
| A | 9.9 | 0.86 | 0.52(12) |
| B | 12.3 | 0.31 | 0.60(13) |
| C | 28.3 | 0.002 | 0.90(12) |
| D | 30.8 | 0.09 | 1.00(13) |
| E | 6.5 | 0.74 | 0.51(15) |
| F | 8.3 | 0.28 | 0.62(16) |

*$Q_{EC}$ values of $^{31}$S, $^{32}$Cl, and $^{32}$Ar—*The $Q_{EC}$ value of $^{31}$S, $Q_{EC} = 5397.99(24)$ keV, deviates slightly from the adopted value $Q_{EC} = 5396.2(15)$ keV [1]. The new $Q_{EC}$ value for this mirror decay changes the $ft$ value from 4798(33) s [5] to 4808(33) s [49] when a $\log_{10} ft$ calculator [49] is used with $T_{1/2} = 2.574(17)$ s [5], branching ratio of 98.837(31)% [5], and the new $Q_{EC}$ value from this Rapid Communication. The corrected $ft$ value for mirror decays $\mathcal{F}t^{\text{mirror}}$ can be calculated with the nucleus-dependent radiative corrections $\delta'_R$ and $\delta^V_{NS}$ and with the isospin-symmetry breaking correction $\delta^V_C$ given in Ref. [5]:

$$\mathcal{F}t^{\text{mirror}} \equiv f_V t\left(1+\delta'_R\right)\left(1+\delta^V_{NS}-\delta^V_C\right) = \frac{2\mathcal{F}t^{0^+\to 0^+}}{\left(1+\frac{f_A}{f_V}\rho^2\right)}, \quad (2)$$

where $f_A/f_V = 1.019\,51$ [5], $\delta'_R = 1.430(29)\%$ [5], and $\delta^V_C - \delta^V_{NS} = 0.79(4)\%$ [5]. The most recent value for $\mathcal{F}t^{0^+\to 0^+}$ is 3071.81(83) s [50]. The new corrected $ft$ value for $^{31}$S is 4839(34) s, which is a little higher but agrees with the value 4828(33) s obtained in Ref. [5]. The new value changes the mixing ratio a little: from $\rho = 0.5167(84)$ [5] to $\rho = 0.5143(84)$. The new value for $\rho$ can be used to calculate standard model values, for example, for the $\beta$-neutrino angular correlation coefficient $a$, the $\beta$ asymmetry parameter $A$, and the neutrino asymmetry parameter $B$.

The revised mass of $^{32}$Cl also changes the $Q_{EC}$ values of $^{32}$Ar and $^{32}$Cl. The new $Q_{EC}$ value of $^{32}$Cl is 12 680.66(65) keV ($^{32}$Cl [this Rapid Communication], $^{32}$S [28]), which is 5.3(66) keV lower than the AME03 value. With the new $^{32}$Cl mass excess value and the values from Refs. [3,42], a new $Q_{EC}$ value for the superallowed $0^+ \to 0^+$ $\beta$ decay of $^{32}$Ar is obtained, $Q_{EC} = 6088.1(20)$ keV. This is in agreement with the result from an IMME fit in Ref. [8], 6087.3(22) keV. The new $Q_{EC}$ value has an effect on the determination of the $\beta - \nu$ angular correlation coefficient $a$ in the superallowed $\beta$ decay of $^{32}$Ar for which a value

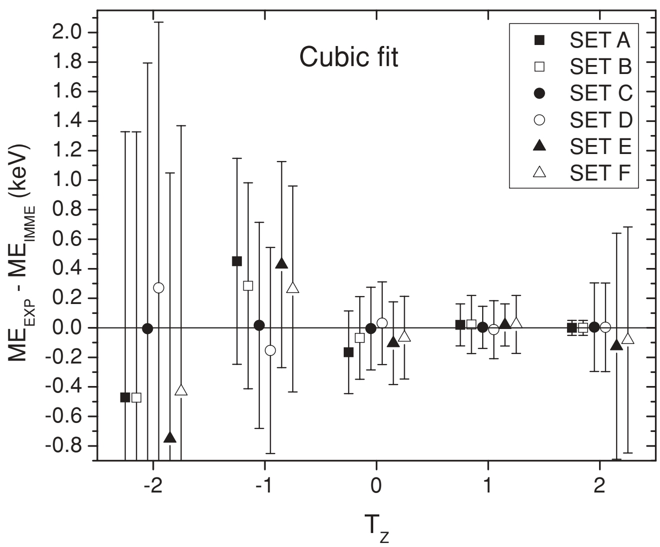


FIG. 5. Differences for the cubic fits of the mass excess values in the $T = 2$ quintet at $A = 32$. The corresponding data sets are given in Table I, and obtained $\chi^2/n$ values and cubic coefficients $d$ are given in Table II. The error bars represent only the uncertainties of the experimental mass excess values.

of $a = 0.9989 \pm 0.0052 \pm 0.0039$(syst) and a dependence of $\partial a/\partial Q = -1.2 \times 10^{-3}$ keV$^{-1}$ were given in Ref. [8]. Therefore, the 0.8-keV change in the $Q_{EC}$ value of the $0^+ \to 0^+$ $\beta$ decay of $^{32}$Ar will shift the value of $a$ for this decay by $-0.0010$.

*Summary and conclusions.* The mass of $^{31}$S has been measured directly and precisely with the JYFLTRAP mass spectrometer. A deviation of 1.4$\sigma$ from the adopted value [1] has been found. The new mass value of $^{31}$S has been used to determine the mass excess of $^{32}$Cl needed for testing the IMME. The quadratic form of IMME has been found to break down, but new direct mass measurements on $^{32}$Si, $^{32}$P, $^{32}$Cl, and $^{32}$Ar are welcome to confirm this. The $Q_{EC}$ value of the mirror nucleus $^{31}$S has been measured, and the corrected $ft$ value has been revised. The $Q_{EC}$ values for $^{32}$Cl and for the superallowed $\beta$ decay of $^{32}$Ar have been updated.

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