High-precision mass measurement of ³¹S with the double Penning trap JYFLTRAP improves the mass value for ³²Cl

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The mass of ³¹S has been measured with the JYFLTRAP double Penning trap mass spectrometer. The new mass excess value of -19042.55(24) keV deviates from the adopted value of -19044.6(15) keV by 1.4σ . The mass value of ³²Cl has been revised with the new ³¹S result and the latest data from β -delayed proton decay of ³²Cl. The new mass excess value for ³²Cl is -13334.88(65) keV, which is the most precise value for ³²Cl so far and in agreement with the recent (³He,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 β decay of T = 1/2 mirror nucleus ³¹S has been determined as $Q_{EC} = 5397.99(24)$ keV, which slightly deviates from the previously adopted value.

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Introduction. ³¹S (Z = 16, N = 15) is a $T_Z = -1/2$ mirror nucleus. Its mass excess (ME), β decay Q_{EC} value, and proton separation energy S_p are essential for several reasons. The adopted ME value of ${}^{31}S$ -19044.6(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 β -delayed proton data [3]. A recent ${}^{32}S({}^{3}He,t){}^{32}Cl$ measurement [4] showed a deviation from the mass value of ³²Cl based on the ³²Cl proton separation energy [3] and the adopted mass of ³¹S [1]. A possible reason for this could be an incorrect ³¹S mass value in Ref. [1]. As the mass of ³²Cl 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 ³¹S.

³¹S β decays to its mirror nucleus ³¹P. 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 ρ [5]. If the mixing ratio is already known, for example, via the determination of the β -neutrino angular correlation $a_{\beta\nu}$, such as for ²¹Na [7], the $|V_{ud}|$ value for the Cabibbo-Kobayashi-Maskawa matrix can be extracted from the corrected *ft* values, the Q_{EC} value has to be known with high precision. In addition to the ³¹S Q_{EC} value, an improved mass value of ³²Cl, and, thus, a more precise Q_{EC} value for ³²Ar, is needed in the studies of positron-neutrino correlations in the superallowed 0⁺ \rightarrow 0⁺ β decay of ³²Ar [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

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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 ³¹S 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/cm² 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 v_c of an ion with a charge q, depends on the magnetic field B and the mass of the ion m_{ion} : $v_c =$ $qB/(2\pi m_{\rm ion})$. 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: $m = (v_c^{\text{ref}} / v_c^{\text{ion}}) \times$ $(m_{\rm ref} - m_e) + m_e$. Here, the obvious choice for the reference ion of 31 S is its β -decay daughter 31 P, which has been measured with very high precision at Florida State University, $m(^{31}P) =$ 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 ³¹S 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 ³¹S file are shown. The total number of ions in the figure is 2002.

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 $\delta_B(v_{\text{ref}})/v_{\text{ref}} = 5.7(8) \times 10^{-11} \text{ min}^{-1} \Delta t$, where Δt 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 ³¹S and ³¹P 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 $\delta_{\rm res,lim}(r)/r = 7.9 \times$ 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 ³¹S and ³²Cl—Altogether 23 measured frequency ratios were obtained, which included 107 585 ions of ³¹S and 99 032 ions of ³¹P 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 ³¹S is -19042.55(24) keV, which deviates by 2.1(15) keV from the adopted value -19044.6(15) keV.

The new mass value agrees with the old β^+ 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).

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

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

$$M(T_Z) = a + bT_Z + cT_Z^2,$$
(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 ³²Cl 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].

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

To test the IMME with the new ³²Cl 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 ³²S, ³²Cl, and ³²Ar are consistent, but the data for ³²Si and ³²P are controversial. There is a huge deviation of 3.2(3) keV(10.6 σ) in the ME of ³²Si, which depends on whether it is taken from the ²⁸Si 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,γ) values of the silicon

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 ³²Si and ³²P 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	-24077.68(30) [34]	0
		E,F	-24 080.86(77) [37]	0
$^{32}\mathbf{P}$	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		-26015.5346(15) [28]	12 047.96(28) [41]
³² Cl	-1		$-13334.64(57)^{d}$	5046.3(4) [3]
³² Ar	-2		-2200.2(18) [42]	0

^aME value of ²⁸Si [18] and S_n values for ²⁹Si-³²Si [1] used. ^bME value of ³¹P [18] and S_n value for ³²P [38,39] used. ^cAn additional systematic error of 20 ppm was taken into account also for Ref. [38] where only a statistical error was given. ^dA 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 χ^2/n values are given in Table II.

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,γ) 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].

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

The LEBIT mass excess value for ³²Si [34] yields the highest χ^2/n values for quadratic fits but surprisingly low χ^2/n values for the cubic fits (see Fig. 5). In fact, the fit with the LEBIT ³²Si [34] value and the ³²P 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 ³²Si and ³²P 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 χ^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	$\chi^2/n_{\rm quadr.}$	$\chi^2/n_{ m cubic}$	d (keV)
A	9.9	0.86	0.52(12)
В	12.3	0.31	0.60(13)
С	28.3	0.002	0.90(12)
D	30.8	0.09	1.00(13)
Е	6.5	0.74	0.51(15)
F	8.3	0.28	0.62(16)

 Q_{EC} values of ³¹S, ³²Cl, and ³²Ar—The Q_{EC} value of ³¹S, $Q_{EC} = 5397.99(24)$ 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 log₁₀ *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 δ'_R and δ^V_{NS} and with the isospin-symmetry breaking correction δ^V_C given in Ref. [5]:

$$\mathcal{F}t^{\text{mirror}} \equiv f_V t \left(1 + \delta_R' \right) \left(1 + \delta_{NS}^V - \delta_C^V \right) = \frac{2\mathcal{F}t^{0^+ \to 0^+}}{\left(1 + \frac{f_A}{f_V}\rho^2 \right)}, \quad (2)$$

where $f_A/f_V = 1.01951$ [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 ³¹S 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 ρ can be used to calculate standard model values, for example, for the β -neutrino angular correlation coefficient a, the β asymmetry parameter A, and the neutrino asymmetry parameter B.

The revised mass of ³²Cl also changes the Q_{EC} values of ³²Ar and ³²Cl. The new Q_{EC} value of ³²Cl is 12 680.66(65) keV (³²Cl [this Rapid Communication], ³²S [28]), which is 5.3(66) keV lower than the AME03 value. With the new ³²Cl mass excess value and the values from Refs. [3,42], a new Q_{EC} value for the superallowed $0^+ \rightarrow 0^+$ β decay of ³²Ar 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 β decay of ³²Ar for which a value

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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 χ^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 $\partial a/\partial Q = -1.2 \times 10^{-3} \text{ keV}^{-1}$ were given in Ref. [8]. Therefore, the 0.8-keV change in the Q_{EC} value of the $0^+ \rightarrow 0^+ \beta$ decay of ³²Ar will shift the value of *a* for this decay by -0.0010.

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

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