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Determination of the Q_{EC} values of the $T = 1/2$ mirror nuclei ^{21}Na and ^{29}P at LEBIT

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We report the first direct measurement of the transition energy Q_{EC} of the ^{21}Na mixed Fermi–Gamow-Teller decay. This is the first of the $T = 1/2$ mirror nuclei decays used for the determination of V_{ud} to be measured with Penning trap mass spectrometry. In addition, the ^{29}P mass was measured directly for the first time and used along with the mass of its daughter, ^{29}Si , for the independent Q_{EC} determination of this decay. The obtained $Q_{EC}(^{21}\text{Na}) = 3547.11(9)$ keV and $\text{ME}(^{29}\text{P}) = -16953.15(47)$ keV significantly improve the latest published values and reduce the contribution of the Q_{EC} uncertainty on $\mathcal{F}t^{\text{mirror}}$ to the same order as the theoretical corrections.

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I. INTRODUCTION

The ongoing search for evidence of physics beyond the standard model is one of the driving forces in fundamental physics research. Particularly crucial for testing the validity of the electroweak model is the unitarity of the Cabibbo-Kobayashi-Maskawa (CKM) matrix that describes the mixing of the different quark generations [1, 2]. The most stringent test is the verification of the first row condition,

$$V_{ud}^2 + V_{us}^2 + V_{ub}^2 = 1 \quad (1)$$

which is dominated by the largest element V_{ud} describing the up-down quark mixing. Values for V_{ud} so far have been obtained from measurements of superallowed $0^+ \rightarrow 0^+$ decays of $T = 1$ nuclei [3], free neutron lifetime measurements [4], pion beta decay rates [5] and beta decay measurements of $T = 1/2$ isospin doublets, also called mirror nuclei [6]. Unlike the $T = 1$ decays which undergo a pure Fermi transition, the decay of the $T = 1/2$ mirror nuclei has a mixed Fermi–Gamow-Teller nature, driven by vector and axial-vector currents [7, 8]. The conservation of vector currents (CVC) is likewise expected for these decays, contrary to the axial-vector current which is not conserved in nuclear transitions. As for the superallowed decays, $\mathcal{F}t^{\text{mirror}}$ is calculated with the vector part of the experimental $f_V t$ value with theoretical corrections, but is not expected to be constant for all nuclei. $\mathcal{F}t^{\text{mirror}}$ can be written as [8]

$$\mathcal{F}t^{\text{mirror}} = f_V t (1 + \delta'_r) (1 + \delta_{\text{NS}}^V - \delta_C^V) \quad (2)$$

with δ'_r and δ_{NS} being the transition-dependent parts of the radiative correction, and δ_C^V , the isospin-symmetry-breaking correction. $f_V t$ is the product of the statistical

rate function, f_V , of the vector part of the decay and the partial half-life, t , of the mother nucleus. Three experimental quantities determine $f_V t$: Q_{EC} , contributing in the fifth power to f_V [9], the half-life, $t_{1/2}$, of the mother nuclide and the branching ratio, BR, of the decay determining t . Half-life and branching ratio contribute proportional and inversely proportional to t , respectively. However, due to the mixed nature of the transition, an additional correction has to be added. The corrected value is given by

$$\mathcal{F}t_0^{\text{mirror}} = \mathcal{F}t^{\text{mirror}} \left(1 + \frac{f_A}{f_V} \rho^2 \right) \quad (3)$$

with f_A denoting the uncorrected statistical decay rate function of the axial-vector part of the decay, and the Fermi to Gamow-Teller mixing ratio, ρ . Provided the CVC hypothesis is correct, this quantity is expected to be isospin independent [10]. Following this, V_{ud} can be deduced for $T = 1/2$ mirror nuclei from [6]

$$V_{ud}^2 = \frac{K}{\mathcal{F}t_0^{\text{mirror}} G_F^2 (1 + \Delta_V^R)} \quad (4)$$

with $K/(\hbar c)^6 = 2\pi^3 \ln 2 \hbar / (m_e c^2)^5 = 8120.271(12) \times 10^{-10} \text{GeV}^{-4}\text{s}$, the Fermi constant $G_F/(\hbar c)^3 = 1.1663787(6) \times 10^{-5} \text{GeV}^{-2}$ and a transition independent correction $\Delta_V^R = 0.02361(38)$ [11].

In the past, experiments mainly focused on the $T = 1$ nuclei and the Q_{EC} value of ^{14}O was the last of the so-called traditional nine to be measured directly with a Penning trap [12]. Presently, the five fully determined $T = 1/2$ mirror nuclei ^{19}Ne , ^{21}Na , ^{29}P , ^{35}Ar and ^{37}K yield the second most precise value for V_{ud} [6]. However, in all cases, experimental uncertainties dominate their $\mathcal{F}t^{\text{mirror}}$ -values [8]. This publication focuses on the improvement of the Q_{EC} value of the ^{21}Na and ^{29}P decays.

The half-life of one of the most precisely known $T = 1/2$

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mirror nuclei, ^{21}Na , has been measured recently [13]. As its Q_{EC} value was only deduced from mass measurements [14–16] we present here the first direct measurement of the Q_{EC} value of the transition $^{21}\text{Na}(\epsilon)^{21}\text{Ne}$ using Penning trap mass spectrometry. We also present the first direct mass measurement of ^{29}P to obtain the Q_{EC} value of the decay $^{29}\text{P}(\epsilon)^{29}\text{Si}$, whose uncertainty is exclusively determined by the $^{28}\text{Si}(p,\gamma)^{29}\text{P}$ reaction [16], while the mass of its daughter nuclide, ^{29}Si , was determined independently with very high precision [17–19].

II. EXPERIMENT DESCRIPTION

The measurements of the Q_{EC} value of the $^{21}\text{Na}(\epsilon)^{21}\text{Ne}$ decay and the mass of ^{29}P were performed at the Low-Energy Beam and Ion Trap (LEBIT) [20] located at the National Superconducting Cyclotron Laboratory (NSCL). LEBIT is unique among the on-line Penning trap mass spectrometers as it is the only device that can perform high-precision mass measurements of radioactive isotopes produced by projectile fragmentation [20]. A $^{36}\text{Ar}^{18+}$ primary beam was accelerated to an energy of 150 MeV/u at the Coupled Cyclotron Facility at the NSCL that was then impinging on a 987 mg/cm² or a 1151 mg/cm² beryllium target for the production of ^{21}Na and ^{29}P , respectively. The beam fragments were sent to the A1900 fragment separator [21] with a 240 mg/cm² aluminum wedge for isotope separation. ^{21}Na was obtained as byproduct in a beam with the A1900 optimized for the separation of ^{24}Si with a rigidity $B\rho = 2.46$ Tm. The only contaminants were ^{20}Ne , ^{22}Mg and ^{23}Al . For ^{29}P the separation was optimized with $B\rho = 2.61$ Tm with only ^{28}Si and ^{30}S in the secondary beam. Next, the secondary beam was decelerated by passing through aluminum degraders. Their thickness was selected so that the beam, after passing a fused silica wedge at the entrance of the gas cell, was decelerated to an energy of less than 1 MeV/u [22]. A schematic overview of the most important components of the low energy beam line used in this experiment is shown in Fig. 1.

Inside the gas cell [22], the ions were stopped by collisions with helium atoms at a pressure of about 93 mbar and recombined to singly- or doubly-charged states. An electric rf field was used to repel the ions from the wall while a combination of electric DC field and gas flow transported them out of the gas cell. The ions were extracted through a radiofrequency quadrupole ion guide (RFQ) from the 30 kV platform towards ground and subsequently separated from non-isobaric stable molecular ion contaminants with a magnetic dipole mass separator having a resolving power of $m/\Delta m \approx 1500$. Reentering the 30 kV platform, the ions were captured in an RFQ cooler and buncher [23] for accumulation and cooling. After being ejected from the cooler/buncher a fast kicker was used as time-of-flight gate, further improving beam

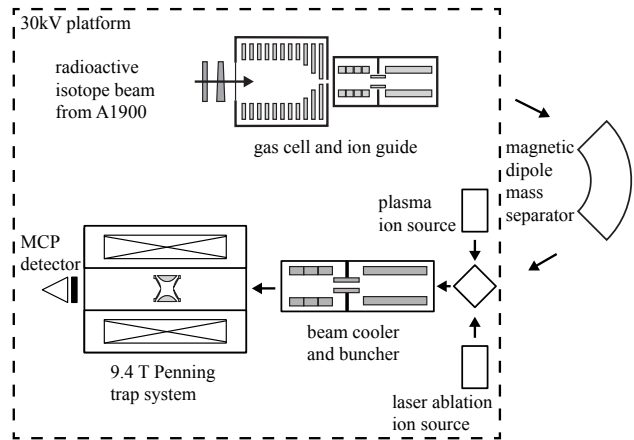


FIG. 1. Schematic overview of the gas cell and LEBIT.

purity.

The hyperbolic Penning trap at the LEBIT experiment is located inside a 9.4 T superconducting magnet. Three dimensional confinement was achieved by a superposition of the strong homogeneous magnetic field \vec{B} and a weak electrostatic quadrupole field \vec{E} with a mixed motion characterized by three eigenmotions, axial, modified cyclotron and magnetron motion with eigenfrequencies ν_z , ν_+ and ν_- respectively [24]. The radius of the latter motion was enlarged by forcing the ions to enter the trap off-axis with a four-fold segmented Lorentz steerer [25]. After capture, the ions were purified by resonantly exciting the modified cyclotron motion of identified contaminants with a dipolar excitation at their modified cyclotron frequency to radii that were large enough to prevent them from interfering with the measurement. The only identified contaminant was $^1\text{H}_3\ ^{18}\text{O}^+$ at $A = 21$, isobaric to $^{21}\text{Na}^+$. In case of ^{29}P , delivered to LEBIT as $(^{29}\text{P}^{40}\text{Ar})^{2+}$ from the gas cell, no contaminants were found as a result of a half-integer mass-to-charge ratio. The mass measurements were carried out via the determination of the free cyclotron frequency of the ion with charge state q and the charge-to-mass ratio qe/m_{ion} , where e is the elementary charge, in the magnetic field with strength B :

$$\nu_c = \frac{1}{2\pi} \frac{qe}{m_{\text{ion}}} B. \quad (5)$$

This frequency was measured using the time-of-flight ion-cyclotron-resonance technique (TOF-ICR) [26, 27], where the conversion from magnetron into modified cyclotron motion is probed with a quadrupolar rf field around the side-band $\nu_{\text{rf}} = \nu_+ + \nu_- = \nu_c$. As radial energy is converted to axial energy in a magnetic field gradient when the ejected ion exits the trap, the change in radial energy in the conversion process is detected as a change in the flight time to a microchannel plate detector (MCP) outside the magnetic field. The mean time of flight is recorded as function of the excitation frequency,

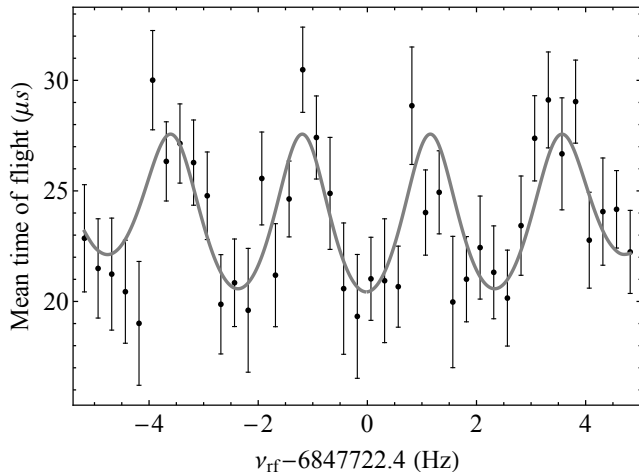


FIG. 2. Example of a Ramsey resonance of $^{21}\text{Na}^+$ with a 100-300-100 ms excitation pattern for the determination of ν_c with the TOF-ICR technique obtained in this work. The solid curve is the fit of the theoretical line shape [30] to the data.

and ν_c is then obtained from a fit of the theoretical line shape to the data points. At LEBIT, the so-called Ramsey technique is applied using a two-pulse excitation separated by a waiting time for a gain in precision compared to the continuous rf excitation of the ion motion [28–30]. A sample resonance of $^{21}\text{Na}^+$ with a 100-300-100 ms excitation scheme is shown in Fig. 2.

In addition to the cyclotron frequency measurements of the ions of interest, the magnetic field strength has to be determined precisely. Thus, the same measurement was performed with a reference ion with a well-known mass prior to and subsequent to the measurement of the ion of interest. The magnetic field was interpolated to the time of the measurement from these two frequencies. The mass of the neutral atom was then derived as

$$m = \frac{\nu_{c,\text{ref}}}{\nu_c} \frac{q}{q_{\text{ref}}} \left(m_{\text{ref}} - q_{\text{ref}} m_e + \frac{\sum_i b_{\text{ref},i}}{c^2} \right) + q m_e - \frac{\sum_i b_i}{c^2}, \quad (6)$$

where $\nu_{c,\text{ref}}$, $(m_{\text{ref}} - q_{\text{ref}} m_e)$ and q_{ref} denote interpolated cyclotron frequency, mass and charge state of the reference ion, $\sum_i b_{\text{ref},i}$ and $\sum_i b_i$ the sum of the first q_{ref} and q ionization energies of the reference ion and the ion of interest, m_e the electron mass and c the speed of light. The carbon cluster $^{12}\text{C}_3^+$, produced by the non-resonant laser ablation ion source, was used as reference for $(^{29}\text{P}^{40}\text{Ar})^{2+}$, while $^{21}\text{Ne}^+$, obtained from natural neon gas ionized in the plasma ion source, was used as the mass reference for $^{21}\text{Na}^+$. Both reference ion sources were located upstream of the cooler buncher (see Fig. 1) and the reference ions were always treated the same as their radioactive counterpart. The Q_{EC} value of the ^{21}Na

decay can then be derived from Eq. (6) as:

$$Q_{\text{EC}} = \left(\frac{\nu_{c,\text{ref}}}{\nu_c} - 1 \right) (m_{\text{ref}} c^2 - m_e c^2 + b_{\text{ref},1}) + b_{\text{ref},1} - b_1. \quad (7)$$

Several effects, such as trap misalignment in the magnetic field, may cause small frequency shifts, thereby shifting the frequency ratio $r = \nu_{c,\text{ref}}/\nu_c$ [31] and therefore the calculated mass of the ion of interest, depending on the charge-to-mass ratio difference to the reference ion. This shift has been determined to be $2.0 \times 10^{-10} q/u$ at LEBIT and is added in quadrature to the statistical uncertainty [32]. Furthermore, non-linear magnetic field fluctuations are known to distort the frequency ratio. However, we found no evidence for such an effect affecting the level of precision reached in this work. The presence of unidentified isobaric contaminants in the trap during the measurement could also lead to frequency shifts [33] which was minimized by ensuring no contaminants were present at a level exceeding a few percent. Also the total number of ions in the trap was limited in the analysis by only considering events with five or fewer ions. With a measured MCP detection efficiency of 63% [12], this corresponds to fewer than nine ions in the trap at the same time.

III. RESULTS AND DISCUSSION

Alternating TOF-ICR measurements of ^{21}Ne and its mother nuclide ^{21}Na were performed with a 100-300-100 ms Ramsey excitation scheme to determine the Q_{EC} value of the ^{21}Na decay. Five frequency ratios containing a total of 3528 $^{21}\text{Na}^+$ ions were recorded yielding a weighted average $r = 1.0001813912(46)$ where only the statistical uncertainty is given. As ^{21}Ne and ^{21}Na form an isobaric doublet, systematic effects due to trap imperfections are negligible at this level of precision. Effects of the mass dependent frequency shift as well as relativistic frequency shifts are negligible as they are both $\Delta\nu_c/\nu_c < 10^{-10}$, more than an order of magnitude below the statistical uncertainty. Furthermore, the Birge ratio [40] of 0.68(21) for the measurement indicates that the uncertainty of the individual measurements was slightly overestimated. Using Eq. (7) with the first ionization energies of ^{21}Ne and ^{21}Na [41, 42] and the ^{21}Ne mass from the Atomic Mass Evaluation 2012 (AME2012) [37] the Q_{EC} value was determined to be 3547.11(9) keV from which the mass excess $\text{ME} = (m - A \times 931494.0023(7) \text{ keV/u}) c^2$ of ^{21}Na , $\text{ME} = -2184.63(10) \text{ keV}$, was deduced with Eq. (6).

The long-lived nuclide ^{29}P was delivered as the weakly bound molecular ion $(^{29}\text{P}^{40}\text{Ar})^{2+}$ from the gas cell, limiting the length of the Ramsey excitation scheme to 50-150-50 ms due to losses by charge exchange with background gas in the Penning trap. However, the cyclotron

TABLE I. Compilation of updated Q_{EC} values, half-lives and branching ratios for ^{19}Ne , ^{21}Na , ^{37}K . Values have been obtained following the procedure in [8], but updated as indicated in the last column.

Nuclide	parameter	Value	Deviations from [8]
^{19}Ne	$t_{1/2}$	17.2585(73) s	updated with [34–36]
	Q_{EC}	3239.49(16) keV	new Q_{EC} deduced [37]
^{21}Na	$t_{1/2}$	22.428(13) s	updated with [13]
	BR	95.253(77)	value from [Ac07] excluded, value from [38] corrected
^{37}K	$t_{1/2}$	1.2363(16)	updated with [39]

TABLE II. Comparison of the Q_{EC} values, f_V , $\mathcal{F}t^{\text{mirror}}$ and the deduced individual V_{ud} for ^{21}Na and ^{29}P . For each nuclide, the first lines show the values from the latest compilation of $T = 1/2$ mirror nuclei [6, 8], the second lines the updated values with the latest updates on the experimental parameters and the third lines show the results including our measurements.

Nuclide	Q_{EC} (keV)	f_V	$\mathcal{F}t^{\text{mirror}}$ (s)	V_{ud}	References
^{21}Na	3547.58(70)	170.97(21)	4085(12)	0.9697(38)	[6, 8]
	3547.14(28)	170.824(84)	4070.1(47)	0.9714(34)	updated Q_{EC} [37], $t_{1/2}$ and BR (see Tab. I)
	3547.11(9)	170.815(27)	4069.9(43)	0.9714(34)	This work
^{29}P	4942.45(60)	1136.7(8)	4809(19)	0.944(44)	[6, 8]
	4942.58(60)	1136.8(8)	4810(19)	0.944(44)	updated Q_{EC} [37]
	4942.18(37)	1136.3(5)	4807(19)	0.945(44)	This work and [37]

frequency of the reference ion $^{12}\text{C}_3^+$ was determined significantly more precisely with a 150-450-150 ms excitation scheme so as not to limit the total precision. From nine frequency ratios with a total of 14433 ($^{29}\text{P}^{40}\text{Ar}$) $^{2+}$ ions a weighted mean of $r = 0.9575574532(70)$ was obtained with a Birge ratio of 0.85(16). Due to the mass-to-charge difference of ($^{29}\text{P}^{40}\text{Ar}$) $^{2+}$ and $^{12}\text{C}_3^+$ the frequency ratio was corrected by -3.1×10^{-10} to account for mass-dependent systematic frequency shifts.

The mass of ^{29}P was obtained from the measured cyclotron frequency ratio of ($^{29}\text{P}^{40}\text{Ar}$) $^{2+}/^{12}\text{C}_3^+$ using Eq. (6), where the binding energies must now include also the molecular binding energies of $^{12}\text{C}_3$ and $^{29}\text{P}^{40}\text{Ar}$. The ionization potential and the binding energy of $^{12}\text{C}_3$ have been determined [43, 44], however both are unknown for $^{29}\text{P}^{40}\text{Ar}$. We assumed that the argon atom just attaches to a doubly charged phosphorus ion with ionization potentials given in [45, 46]. Furthermore, we added the difference between the first two ionization potentials of ^{29}P and ^{40}Ar [47, 48] of 12.9 eV in quadrature as an additional uncertainty. Finally, the mass of ^{40}Ar is subtracted from the determined molecular mass assuming that the molecular binding energy with a noble gas is negligible on this level of precision, yielding a mass excess of $\text{ME} = -16953.15(47)$ keV. The error is dominated by the statistical uncertainty at more than 95 %. As the mass excess published in the AME2012, $\text{ME}_{\text{AME}} = -16952.5(60)$ keV, is almost exclusively determined by one measurement of the reaction $^{28}\text{Si}(p, \gamma)^{29}\text{P}$ the weighted average was calculated to be $\text{ME}_{\text{av}} = -16952.90(37)$ keV, from which $Q_{\text{EC}} = 4942.18(37)$ keV was derived for the decay $^{29}\text{P}(\epsilon)^{29}\text{Si}$.

With the newly determined Q_{EC} values, $\mathcal{F}t^{\text{mirror}}$ and V_{ud} for ^{21}Na and ^{29}P were calculated using Eq. (2). The

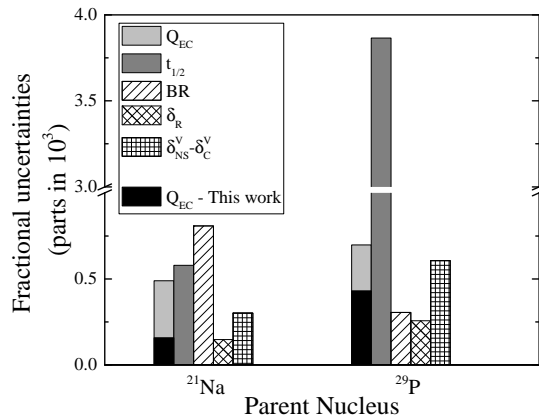


FIG. 3. Fractional uncertainties in $\mathcal{F}t^{\text{mirror}}$ of all contributing experimental (Q_{EC} (released energy in the nuclear decay), $t_{1/2}$ (half-life of the mother nuclide), BR (branching ratio of the decay)) and theoretical parameters (δ_r' (radiative correction from QED), $\delta_{\text{NS}}^V - \delta_c^V$ (difference of nuclear structure and isospin symmetry breaking correction)).

values for the theoretical corrections, δ_r' and $\delta_{\text{NS}}^V - \delta_c^V$, and for the electron capture probability, entering the partial half-life t as a third parameter, were taken from [8] as well as half-life and branching ratio for ^{29}P . New averages were calculated for half-life and branching ratio of ^{21}Na following the procedure established in [8] (see Tab. I), involving the exclusion of data with an uncertainty at least ten times larger than the most precise measurement. The value from the private communication [Ac07] in [8] is not considered for the calculation of the average branching

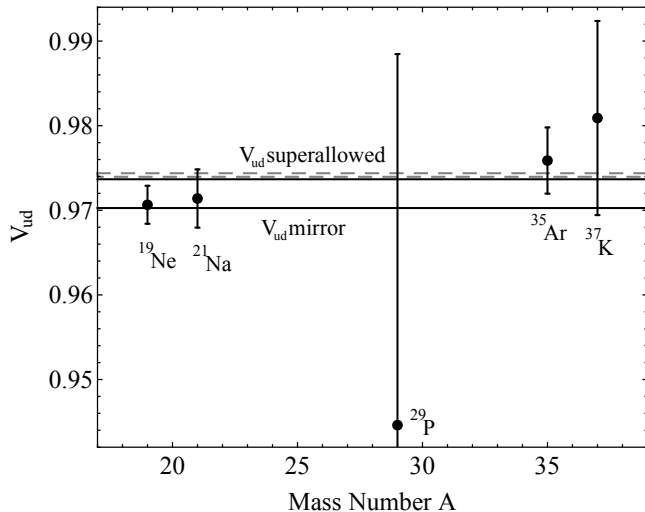


FIG. 4. Updated V_{ud} values for the $T = 1/2$ mirror nuclei. The individual values were calculated as described in [6, 8] using the updated averages (see Tab. I) and the Q_{EC} values obtained in this work (see Tab. II). The solid lines mark the 1- σ uncertainty of the average value. For comparison the 1- σ boundary of V_{ud} deduced from superallowed β decays are shown as dashed lines [3].

ratio, as the same authors published a significantly different and less precise value [49]. In addition, the branching ratio extracted from [38] ([Wi80] in [8]) was corrected to 95.03(16)% as the authors of [8] referred to a weighted average calculated in [38] instead of the measurement result.

The vector part of the statistical rate functions f_V were calculated with the new Q_{EC} values using the parametrization presented in [9]. These results are given in Table II along with $\mathcal{F}t^{\text{mirror}}$. An overview on the fractional uncertainties of all contributing parameters is shown in Fig. 3. The ratio of the statistical rate functions, f_A/f_V , was also recalculated with the parametrization [9] but remained the same within the present level of precision. With Eqs. (3) and (4) as well as ρ extracted from [6] the individual values for V_{ud} are derived. They are displayed in Fig. 4 together with updated values from all five $T = 1/2$ mirror nuclei with an experimentally determined ρ using the newly determined averages shown in Table I.

The uncertainties of the Q_{EC} values and thereby f_V of both nuclides, ^{21}Na and ^{29}P , were reduced significantly compared to the previous measurements. Their contribution to the error budget of $\mathcal{F}t^{\text{mirror}}$ was reduced to the same level as the theoretical corrections (see Fig. 3). However, the improvement in Q_{EC} did not affect $\mathcal{F}t^{\text{mirror}}$ for ^{29}P and only marginally for ^{21}Na , respectively. As

depicted in Fig. 3, this is due to other dominating uncertainties, the half-life for ^{29}P and the branching ratio for ^{21}Na . Due to the large uncertainty in ρ , V_{ud} remains unchanged in both cases.

As the updated $\mathcal{F}t^{\text{mirror}}$ of ^{21}Na deviates from the latest published value [13], it should be pointed out that the Q_{EC} value was deduced from the masses published in the AME2003 [50], so that the uncertainty of the branching ratio and not the Q_{EC} value is dominating the uncertainty of $\mathcal{F}t^{\text{mirror}}$. Therefore, the average for V_{ud} was re-determined using updated values of the five nuclei shown in Fig. 4 yielding:

$$\bar{V}_{ud} = 0.9720(17) . \quad (8)$$

This value is close to the one so far reported for $T = 1/2$ mirror nuclei [6] and deviates only by 1.3 σ from $V_{ud}^{0^+ \rightarrow 0^+} = 0.97417(21)$ derived from superallowed β decays [3]. Thus, further measurements are required for the Gamow-Teller to Fermi mixing ratios ρ , as their uncertainties surpass the others in all cases significantly or are not even derived from experiments. Further half-life measurements are also required, as their uncertainties are in some cases the limiting factor and, as shown for example in [13, 34], independent measurements rarely yield the same value.

IV. CONCLUSION

The first direct high-precision measurements of the Q_{EC} value of ^{21}Na and the mass of ^{29}P have been performed with the aim to improve the $\mathcal{F}t^{\text{mirror}}$ values used to deduce V_{ud} . From these measurements $Q_{\text{EC}}(^{21}\text{Na}) = 3547.11(9)$ keV and $Q_{\text{EC}}(^{29}\text{P}) = 4942.18(37)$ keV were obtained, improving the previously published values by factors of three and 1.5, respectively. The contribution to the total uncertainty of $\mathcal{F}t^{\text{mirror}}$ and V_{ud} is now at the same level as the theoretical corrections. With these improvements $V_{ud} = 0.9720(17)$ has been calculated for $T = 1/2$ mirror nuclei which is agreement with the value presented in the most recent review [6].

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