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Improved high-precision mass measurements of mid-shell neon isotopes

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Abstract

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New mass values measured with the TITAN MR-TOF-MS are reported for the short-lived isotopes $^{24-26}\text{Ne}$, produced at TRIUMF's ISAC facility using a uranium carbide target and cold FEBIAD ion source. A least-squares adjustment using the program which produces the Atomic Mass Evaluation was performed and the improved precision of the new mass values is highlighted. The improved mass accuracy in this $N = 14$ mid-shell region can also point to the refinement in values of the charge radii. By reducing the mass uncertainty of isotopes, their contribution of the mass shift uncertainty in laser spectroscopy can be negated. The work is part of developments for reaching the $N = 20$ Island of Inversion, where high molecular contamination hinders mass measurements of the neon isotopes.

Keywords: mass spectrometry, ion trapping, mass evaluation, nuclear structure

1. Introduction

One key property of all nuclei is their mass. Importantly, the mass of a nucleus is not solely dependent on its constituent parts, protons and neutrons, but also the forces within it. When considered as a whole, these forces amount to a single quantity referred to as the binding energy. Due to the analogous nature of mass and energy, this binding energy causes a shift in the mass of the nucleus such that the final atomic mass is:

$$m(A, Z) = m_p Z + m_n(A - Z) + m_e Z - E_{B_n}/c^2 - E_{B_e}/c^2 \quad (1)$$

where A is nucleon number, Z is proton number, m_p is proton mass, m_n is neutron mass, m_e is electron mass, E_{B_n} is the nuclear binding energy, and E_{B_e} is electron binding energy.

A prominent description for the organization of these binding forces is the nuclear shell model (1), in which the magic numbers: 2, 8, 20, 28, 50, 82, and 126 arise and are indicative of large energy gaps between nucleon orbitals that increase nuclear stability. However, as experiments push further from stability and reach greater proton-neutron asymmetry, shell effects can diminish and even disappear altogether (2).

One of the most prominent examples of this is the $N = 20$ Island of Inversion, discovered by mass measurements that showed the nuclear binding energy to increase instead of decrease from ^{31}Na to ^{32}Na ($N = 20$ and 21, respectively) (3). Many other experimental observables have also been studied for $N = 20$ and other erstwhile shell closures. For a review see (4).

22 Since the initial Island of Inversion findings, there have been several high
23 precision mass spectrometry campaigns to map the Island of Inversion bor-
24 ders. Many of those have been pursued using the TITAN spectrometer (5) at
25 TRIUMF's ISAC facility for radioactive ion beams (6; 7; 8). This paper de-
26 scribes our first attempt to address the isotopes of the neon ($Z = 10$) chain.
27 ISAC is a thick-target, ISOL facility from which the radioactive species dif-
28 fuse as atoms. Since neon is a noble gas, it must be ionized using a discharge
29 source, which unfortunately ionizes many other species in the process. Com-
30 pounding the problem is the ionization of molecular species formed in the
31 hot environment, causing copious background. In this first attempt, only the
32 isotopes $^{24-26}\text{Ne}$ could be measured. To go further, a scheme for breaking
33 up the molecules forming the background has been developed, which is the
34 subject of a separate publication (9). Here we present the mass measurement
35 results.

36 2. The TITAN Experimental Facility

37 The neon isotopes measured in this experiment were produced at the Iso-
38 tope Separator and ACcelerator (ISAC) (10) facility located at TRIUMF in
39 Vancouver, Canada. The primary beam of 480 MeV protons with a current
40 of $9.8\mu\text{A}$ impinged on a UCx target. The neutral product isotopes then dif-
41 fused out of the target and through a cold transfer line. They were then
42 ionized using a Forced Electron Beam Arc Discharge (FEBIAD) ion source
43 to create the secondary radioactive ion beam (RIB) (11). The combination
44 of ion source and cold transfer line was selected due to the high first ion-
45 ization potential of Ne as well as the expected presence of large amounts of
46 background species with much lower first ionization potentials (e.g. Na and
47 Mg). Ultimately, the cold transfer line was successful in reducing the rate of
48 Na and Mg relative to Ne by many orders of magnitude.

49 The RIB is reduced to a single mass unit (i.e. selects for a single A/q) via
50 two dipole mass separators and is then delivered to TRIUMF's Ion Trap for
51 Atomic and Nuclear science (TITAN) (5). The continuous beam from ISAC
52 is accumulated and cooled first in the helium buffer gas filled RFQ cooler
53 and buncher (12) which is set to a potential to match the incoming ≈ 20 keV
54 beam energy. After accumulating for 10 ms, the cold ion bunch is ejected
55 towards the Multi-Reflection Time-of-Flight Mass-Spectrometer (MR-TOF-
56 MS) (13). Between the two traps, the ions pass through a pulsed drift tube
57 which reduces the beam's energy to match that of the MR-TOF-MS's input

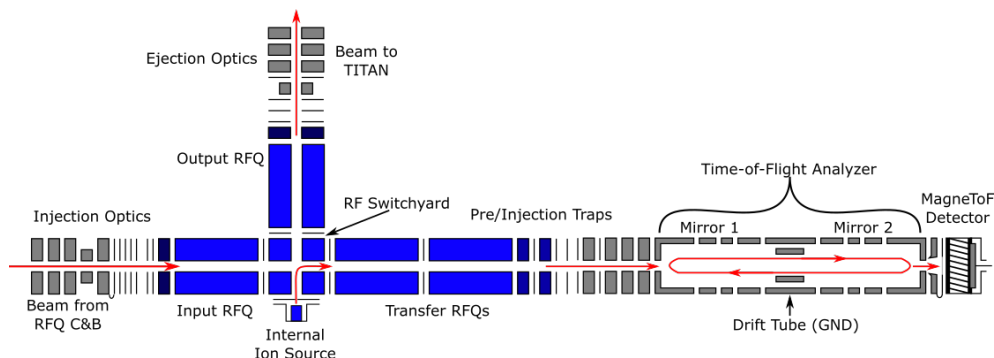


Figure 1: Illustration of the TITAN MR-TOF-MS used to measure the masses in this work. For details, see text.

58 RFQ (≈ 1.4 keV). The ion bunch is captured in the input RFQ by switching
 59 the potential of an entrance aperture from 1.3 kV (transmission) to 1.5 kV
 60 (blocking) once the ion bunch is in the RFQ. This allows for additional A/q
 61 selection from the TITAN RFQ cooler and buncher to negate any molecular
 62 effects due to contamination in the He buffer gas such as charge exchange
 63 and molecular adduction. These molecular reactions have been identified
 64 with off-line stable molecular beam experiments such as performed in (9).

65 A schematic of the TITAN MR-TOF-MS is shown in figure 1 (a detailed
 66 description is given in (14)). To perform a mass measurement, the
 67 captured ions are transported through a six-way RF switchyard (15) and a
 68 series of RFQs before undergoing a final cooling stage in the RF injection
 69 trap. The ions are then injected into the time-of-flight mass analyzer (16).
 70 To confine the ions, the electric potential of the first ion mirror is raised fol-
 71 lowing injection. The ions are then reflected for a set number of turns before
 72 the electric potential of the second mirror is lowered and they impinge on a
 73 Time-of-Flight (TOF) detector (ETP MagneTOF DM572). The signal from the
 74 detector is then converted and recorded using a time-to-digital converter
 75 (FastCom TDC MCX6A) to generate a TOF spectrum.

76 Alternatively, ions can be extracted from the analyzer section by lowering
 77 the potential of the first mirror. The ions can then travel back into the
 78 injection trap which, when closed at a predetermined time, can recapture a
 79 particular species of interest while disposing of the other background species.
 80 This process, referred to as mass-selective re-trapping, is a well established
 81 technique at the TITAN MR-TOF-MS and enables a background suppression
 82 of up to four orders of magnitude with a separation power (the re-trapping

83 analog to mass resolving power) of 100,000 (17; 18). As a result, a dynamical
84 time focus shift is needed to reach high resolving powers for both the re-
85 trapping and mass measurement cycles, particularly when operated with a
86 different number of turns (19).

87 Following re-trapping, the ions can be transported back down the MR-
88 TOF-MS RFQs and out to the rest of TITAN or re-injected into the TOF
89 analyzer where a mass measurement will be performed. As such, the TITAN
90 MR-TOF-MS can act as its own isobar separator. An example of a TOF
91 spectrum before and after re-trapping was employed can be seen in top panel
92 of figure 2. Initially, the beam composition is dominated by $^{24}\text{Mg}^+$ and $^{12}\text{C}_2^+$.
93 However, with re-trapping, the relative composition of $^{24}\text{Mg}^+$ and $^{24}\text{Ne}^+$
94 shifts by four orders of magnitude while the relative composition of $^{12}\text{C}_2^+$ and
95 $^{24}\text{Ne}^+$ shifts by two orders of magnitude. This was done by adjusting the
96 capture time of the trap to ensure there would still be sufficient statistics of
97 $^{12}\text{C}_2^+$ to perform a calibration. As a result of re-trapping, mass measurements
98 were able to be performed with an isotope of interest to background ratio
99 of $1 : 10^4$, $1 : 10^5$, and $1 : 10^6$ for ^{24}Ne , ^{25}Ne , and ^{26}Ne respectively. The
100 resulting mass spectra can be seen in figure 2 with all species identified.

101 In this particular experiment, the RFQ cooler and buncher and MR-
102 TOF-MS were operated with a 100 Hz cycle frequency (instead of a more
103 common 30-50 Hz) to accommodate the short half-lives and to reduce the
104 charge-exchange probability in the He buffer-gas-filled RFQs. To accomplish
105 this, two adjustments were needed. First, because a change in switching
106 frequency changes the voltage drop across the HV switches, the analyzer
107 voltages must be re-tuned whenever the cycle frequency is changed. The
108 voltages which define the reflection potential are sensitive to ≈ 10 mV. When
109 the 100 Hz and 50 Hz tunes are compared, a difference in 10 V can be needed
110 to properly readjust the voltages. Second, because the cycle times are shorter
111 than normal, the drag fields of the MR-TOF-MS RFQs needed to be adjusted
112 to ensure all ions travel from the input RFQ to the pre-trap in 2 to 3 ms.
113 As a result of the shorter cycle times, the ion bunches would fly in the TOF
114 analyzer for 380 to 440 turns which corresponds to $\approx 3.7 - 4.4$ ms. Due to
115 this shorter flight path, the achieved mass resolving power for this experiment
116 was $\approx 200,000$. Additionally, re-trapping was consistently employed due to
117 the large amount of background. Lastly, the resonant frequency of the RF
118 circuit for the RFQs was boosted from 1.11 MHz to 1.67 MHz to better
119 enable transport of lighter ionic species as described in (14).

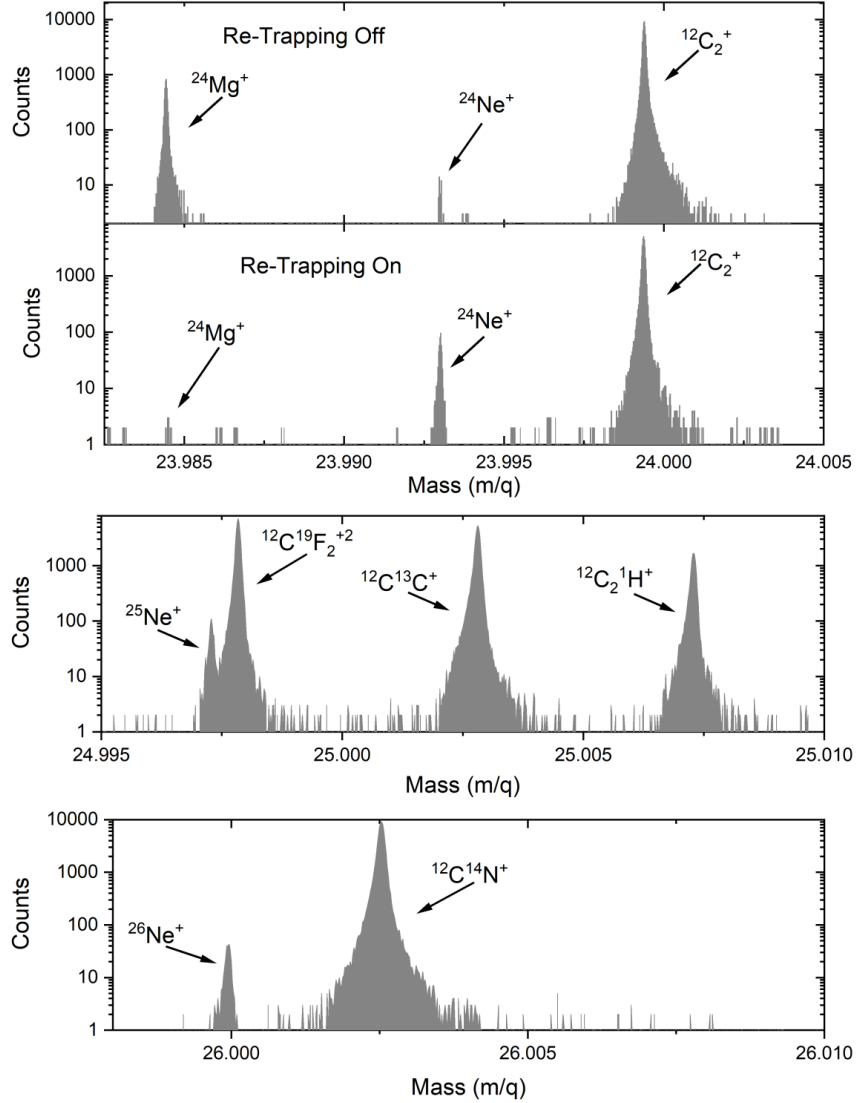


Figure 2: (Top) A comparison of RIB spectra at $A/q = 24$ with re-trapping on and off. The specific timing for re-trapping in this case was chosen such that the $^{24}\text{Mg}^+$ was maximally suppressed while enough $^{12}\text{C}_2^+$ remained to perform a mass calibration. The operation of the MR-TOF-MS allowed for a resolving power of $\approx 200,000$ with $^{24}\text{Ne}^+$ and $^{12}\text{C}_2^+$ separated by ≈ 500 ns in time of flight. (Middle) The RIB spectrum at $A/q = 25$ with all species identified with re-trapping enabled. (Bottom) The RIB spectrum at $A/q = 26$ with all species identified with re-trapping enabled.

120 **3. Data Analysis**

121 In general, the data analysis procedure followed the techniques described
 122 in (20; 21; 22). Using the home-built Mass Acquisition (MAc) software pack-
 123 age (23; 24), the TOF can be converted to mass using:

$$m = c \cdot \frac{(t + t_{\text{start}} - t_0)^2}{(1 + b \cdot T)^2} \quad (2)$$

124 where c , t_0 , and b are calibration parameters, t_{start} is the calculated delay
 125 time before triggering the TDC, T is the number of turns done by a species,
 126 and t is the measured TOF from the opening of the second mirror. To
 127 calibrate the spectrum, t_0 and c are determined using several species spanning
 128 multiple mass units before an experiment. In this particular experiment
 129 $^{27}\text{Al}^+$, $^{28}\text{Si}^+$, $^{39}\text{K}^+$, and $^{41}\text{K}^+$ were used. This allows for b to then be used
 130 as the on-line calibration parameter and enables the calibration species to
 131 be non-isobaric when accounting for the number of turns, T . In most cases,
 132 T for the calibration species and the species of interest is the same, and the
 133 formula above can be simplified to: $m = c(t + t_{\text{start}} - t_0)^2$.

134 After converting a spectrum from TOF to mass, the histogram is ex-
 135 ported and fitted using an open-source Python package (25) in which a
 136 hyper-exponentially modified Gaussian line shape is used (26). After the
 137 centroids of all peaks are determined, a final calibration using

$$\frac{m}{m_{\text{cal}}} = \frac{m_{\text{fit}}}{m_{\text{cal,fit}}} \quad (3)$$

138 is performed to extract the mass values of each species in the spectrum. This
 139 calculation relates the ratio of the fits for the species of interest (m_{fit}) and
 140 the calibrant ($m_{\text{cal,fit}}$) to the literature calibrant mass (m_{cal}) and final mass
 141 (m) of the species of interest.

142 As mentioned above, spectra were taken with a different number of turns
 143 for each isobar requested from ISAC. Each spectrum underwent an inde-
 144 pendent fitting analysis, and a weighted mean was taken to obtain a final
 145 value. In normal circumstances, a systematic uncertainty is determined us-
 146 ing off-line stable-beam tests before and after an experiment. The dominant
 147 contribution to the systematic uncertainty was the non-ideal electrical fields
 148 during extraction of ions from the analyzer. To test the extent of this effect,
 149 the timing for the lowering of the potential of the second electrostatic mir-
 150 ror was scanned and determined the relative systematic uncertainty to be

Species	Calibrant	$\frac{m}{m_{cal}}$	ME _{TITAN} (keV/c ²)	ME _{AME} (keV/c ²)	ME _{AME2020} (keV/c ²)
²⁴ Ne	¹² C ₂	0.99973367 (31)	-5954.0 (69)	-5951.6 (5)	-5951.6 (5)
²⁵ Ne	¹² C ¹³ C	0.99977893 (34)	-2023.8 (80)	-2023.5 (78)	-2036 (29)
²⁶ Ne	¹² C ¹⁴ N	0.99990063 (33)	456.4 (80)	460.4 (74)	481 (18)

Table 1: Ne mass measurements with their respective calibrants, mass ratios, and mass excess TITAN results (ME_{TITAN}). Additionally, values after AME evaluation with the new TITAN results (ME_{AME}) and AME2020 (29) values are shown. All species were measured in the singly charged (1+) state. Mass Excesses and mass ratios are reported as atomic masses.

151 $\delta m_{sys}/m = 3 \times 10^{-7}$. This value is in agreement with previously reported
152 systematic uncertainties as seen in (27; 28).

153 A final precaution is taken for relatively light species measured using
154 the TITAN MR-TOF-MS. Because the ion mirror must be switched open to
155 extract the ions, only a part of a total ion cycle is usable ($\approx 60\text{-}70\%$). For
156 light species, the absolute time window for good measurements becomes very
157 small ($\approx 9 \mu\text{s}$ for $A/q = 24$), and, as a result, the single turn time must be
158 set with a high degree of precision. To determine this window, an off-line
159 scan is done of the opening time of the analyzer to find the region in which
160 the time of flight of the ions is undisturbed (see (20) for a full description
161 of the procedure). To double check this precision, an on-line test measuring
162 the mass of ²⁴Mg⁺ using ¹²C₂⁺ as a calibrant at a similar number of turns
163 to the subsequent mass measurements while the opening time of the second
164 ion mirror was adjusted confirmed that we correctly set the single turn time.
165 This was confirmed due to the test measurements agreeing with literature
166 values within statistical uncertainty ($\delta m/m \approx 10^{-7}$).

167 4. Results and Discussion

168 Table 1 lists the mass values derived for ^{24–26}Ne, as well as the calibrant
169 ion and corresponding mass ratios. This is reported to accommodate for any
170 eventual changes in calibrant-ion mass. The ratios were input to the Atomic
171 Mass Evaluation, which links all data from mass measurement performed by
172 either mass doublets or by reactions or decays, which can be compared for
173 consistency. This combinatorial evaluation procedure produces the atomic
174 mass table. The new TITAN masses are compared to the literature data in
175 figure 3. Here we describe these different measurements that the AME has
176 linked to provide a final evaluated value.

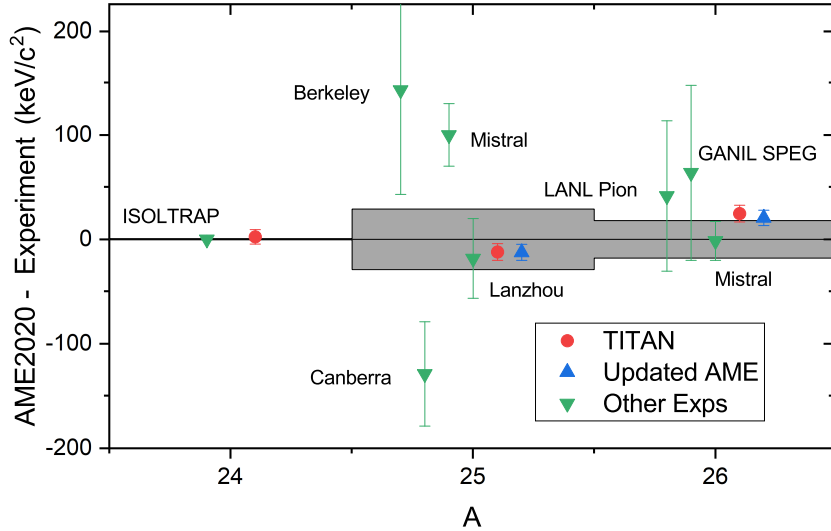


Figure 3: A comparison of the different measurements for ^{24}Ne , ^{25}Ne and ^{26}Ne with the new TITAN results relative to the AME2020. Additionally, the new evaluated masses with the TITAN results are included. The uncertainty in the AME2020 value is shown in gray.

177 The mass of ^{24}Ne ($T_{1/2} = 203$ s) was measured by ISOLTRAP (30) using
 178 the TOF-ICR method in a Penning trap. The ISOLTRAP value is thirteen
 179 times more precise than the new TITAN MR-TOF-MS result but the two
 180 values are in excellent agreement. While the new result is given no statistical
 181 weight in the AME calculation, the value is still used for a consistency check
 182 and slightly strengthens the accuracy for this mass.

183 The ^{25}Ne ($T_{1/2} = 602$ ms) mass has been determined via Q values of
 184 the reactions $^{26}\text{Mg}(^7\text{Li}, ^8\text{B})^{25}\text{Ne}$ at Berkeley (31) and $^{26}\text{Mg}(^{13}\text{C}, ^{14}\text{O})^{25}\text{Ne}$ at
 185 Canberra (32), by direct mass spectrometry with MISTRAL (33; 34), and
 186 by the CSR storage ring at Lanzhou (35). Although the reaction values
 187 showed a roughly 2σ deviation, they were averaged in the AME, yielding
 188 a mass compatible with our measurement, as is the one from the Lanzhou
 189 storage ring. The MISTRAL value deviated by 3.4σ from the evaluated mass
 190 so was rejected from the AME since the mass spectrum showed a double
 191 peak. Coincidentally, the TITAN MR-TOF-MS was able to resolve a near
 192 lying double peak caused by $^{12}\text{C}^{19}\text{F}_2^{2+}$ which could have been the cause of
 193 the previously reported double peak. The TITAN MR-TOF-MS reached a
 194 resolving power of $\approx 200,000$ in this experiment what the required resolving

195 power to separate these two species is $\approx 40,000$. The TITAN result now
 196 accounts for 93% of the AME value.

197 The mass of ^{26}Ne ($T_{1/2} = 197$ ms) has been determined via Q value of the
 198 pion-exchange reaction $^{26}\text{Mg}(\pi^-, \pi^+)^{26}\text{Ne}$ at Los Alamos (36) and directly
 199 by magnetic rigidity combined with time of flight using SPEG at GANIL
 200 (37) and the RF spectrometer MISTRAL (33; 34). All of the data are in
 201 agreement but the superior precision of the new TITAN value accounts 83%
 202 of the new evaluated mass.

203 Nuclear structure effects can be nicely revealed using both the two-neutron
 204 separation energy (S_{2n}):

$$S_{2N}(N, Z) = [m(N - 2, Z) - m(N, Z) + 2m_n] \cdot c^2 \quad (4)$$

205 and the empirical shell gap (ΔS_{2N}):

$$\Delta S_{2N}(N, Z) = S_{2N}(N, Z) - S_{2N}(N + 2, Z) \quad (5)$$

206 The S_{2n} and ΔS_{2N} values for the neon isotopes calculated from the AME (29)
 207 and the new values are plotted in figure 4. While the values have changed
 208 very little with the new, improved, masses, they are now more accurate
 209 due to the good agreement with previous data. Additionally, the S_{2n} and
 210 ΔS_{2N} from different mass models are compared to the experimental data.
 211 In general, the models accurately recreate the trends seen in both the S_{2n}
 212 and ΔS_{2N} , particularly, a local minimum in ΔS_{2N} at $N = 12$ or 13 and local
 213 maximum at $N = 15$.

214 Figure 5 superimposes both the empirical shell gap as well as the differ-
 215 ence in mean-square charge radii, obtained from laser spectroscopy (41) along
 216 the neon isotopic chain. The ΔS_{2n} values reflect the gain in binding energy
 217 from the $d_{5/2}$ sub-shell closure at $N = 15$ whereas the charge radius is lowest
 218 for $N = 14$, raising the question of a new magic number, in place of $N = 20$.
 219 This point is discussed by Marinova et al. (41), who also included the re-
 220 sults of microscopic Fermionic Molecular Dynamics (FMD) calculations that
 221 include contributions from clustering from work on the lighter neon isotopes
 222 (42). As mentioned in both laser spectroscopy papers (41; 42), a source of
 223 systematic uncertainty in the charge radii is from the mass shift as depicted
 224 in figure 5. However, for high precision mass measurements, the contribution
 225 to the final uncertainty is $\approx 0.01\%$ (42). Thus, the new mass data will also
 226 reduce the uncertainties of these observables to better refine the transition

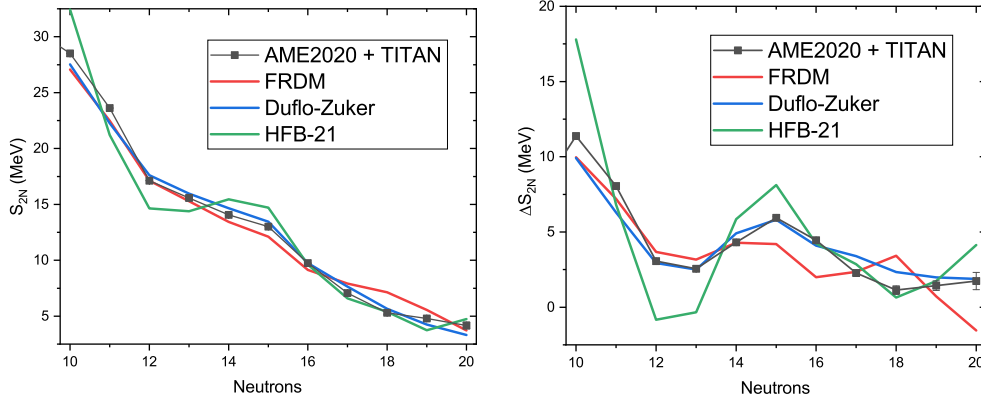


Figure 4: The two-neutron separation energy (S_{2n} , Left) and empirical shell gap (ΔS_{2N} , Right) for the chain of ($Z = 10$) neon isotopes (calculated using the new masses combined with those from the AME2020 (29)). The new experimental data is combined with mean-field calculations using FRDM (38), Duflo-Zuker (39), and HFB-21 (40).

227 towards the $N = 20$ Island of Inversion, and serves as a motivating factor to
 228 perform high precision mass measurements beyond ^{26}Ne .

229 5. Conclusion

230 New mass values for the short-lived $^{24-26}\text{Ne}$ isotopes are reported and a
 231 mass evaluation was performed. The value for ^{24}Ne is in good agreement with
 232 a more precise Penning-trap value (30) while the precisions for the heavier
 233 two were improved by factors of more than three and two, respectively. The
 234 new precision mass values can eventually help reduce the uncertainties of
 235 the charge radii for these isotopes (41). It would be interesting to extend
 236 the FMD calculations from the lighter neon isotopes (42) to see if cluster
 237 configurations continue to account for some of the binding energy. Since the
 238 experimental campaign described in this work, several technical initiatives
 239 have been undertaken to extend the reach of the TITAN MR-TOF-MS into
 240 the $N = 20$ Island of Inversion with Ne including the development of collision-
 241 induced dissociation (43; 9) and improvements to He buffer gas quality.

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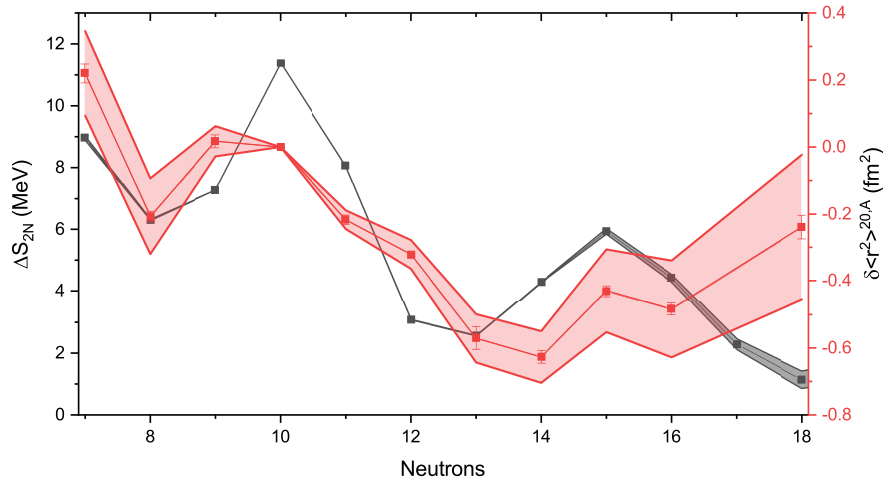


Figure 5: The empirical shell gap (ΔS_{2n}) for the chain of ($Z = 10$) neon isotopes (calculated using the new masses combined with those from the AME2020 (29)), superimposed with the mean-square charge radii from (41). The statistical uncertainty is shown via red error bars while they systematic uncertainty, resulting largely from unknown mass shifts, is shown by the lightly shaded red area.

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