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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 $^{24-26}$ 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 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:

$$m(A,Z) = m_p Z + m_n (A - Z) + m_e Z - E_{B_n} / c^2 - E_{B_e} / c^2$$
(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 ³¹Na to ³²Na (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).

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

³⁶ 2. The TITAN Experimental Facility

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

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



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

⁵⁸ RFQ ($\approx 1.4 \text{ keV}$). 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).

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

Alternatively, ions can be extracted from the analyzer section by lowering the potential of the first mirror. The ions can then travel back into the injection trap which, when closed at a predetermined time, can recapture a particular species of interest while disposing of the other background species. This process, referred to as mass-selective re-trapping, is a well established technique at the TITAN MR-TOF-MS and enables a background suppression of up to four orders of magnitude with a separation power (the re-trapping analog to mass resolving power) of 100,000 (17; 18). As a result, a dynamical
time focus shift is needed to reach high resolving powers for both the retrapping and mass measurement cycles, particularly when operated with a
different number of turns (19).

Following re-trapping, the ions can be transported back down the MR-87 TOF-MS RFQs and out to the rest of TITAN or re-injected into the TOF 88 analyzer where a mass measurement will be performed. As such, the TITAN 89 MR-TOF-MS can act as its own isobar separator. An example of a TOF 90 spectrum before and after re-trapping was employed can be seen in top panel 91 of figure 2. Initially, the beam composition is dominated by ${}^{24}Mg^+$ and ${}^{12}C_2^+$. 92 However, with re-trapping, the relative composition of ${}^{24}Mg^+$ and ${}^{24}Ne^+$ 93 shifts by four orders of magnitude while the relative composition of ${}^{12}C_2^+$ and 94 ²⁴Ne⁺ shifts by two orders of magnitude. This was done by adjusting the 95 capture time of the trap to ensure there would still be sufficient statistics of 96 $^{12}C_2^+$ to perform a calibration. As a result of re-trapping, mass measurements 97 were able to be performed with an isotope of interest to background ratio 98 of $1 : 10^4$, $1 : 10^5$, and $1 : 10^6$ for ²⁴Ne, ²⁵Ne, and ²⁶Ne respectively. The 99 resulting mass spectra can be seen in figure 2 with all species identified. 100

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



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 ²⁴Mg⁺ was maximally suppressed while enough ¹²C₂⁺ remained to perform a mass calibration. The operation of the MR-TOF-MS allowed for a resolving power of $\approx 200,000$ with ²⁴Ne⁺ and ¹²C₂⁺ 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

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

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

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

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

$$\frac{m}{m_{\rm cal}} = \frac{m_{\rm fit}}{m_{\rm cal, fit}} \tag{3}$$

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

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

Species	Calibrant	$\frac{m}{m_{cal}}$	$\rm ME_{TITAN}~(keV/c^2)$	${\rm ME}_{AME}~({\rm keV/c^2})$	$\rm ME_{AME2020}~(keV/c^2)$
²⁴ Ne	$^{12}C_2$	0.99973367(31)	-5954.0 (69)	-5951.6(5)	-5951.6(5)
$^{25}\mathrm{Ne}$	$^{12}\mathrm{C}^{13}\mathrm{C}$	$0.99977893\ (34)$	-2023.8 (80)	-2023.5(78)	-2036 (29)
²⁶ Ne	$^{12}\mathrm{C}^{14}\mathrm{N}$	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.

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

A final precaution is taken for relatively light species measured using 153 the TITAN MR-TOF-MS. Because the ion mirror must be switched open to 154 extract the ions, only a part of a total ion cycle is usable ($\approx 60-70\%$). For 155 light species, the absolute time window for good measurements becomes very 156 small ($\approx 9 \ \mu s$ for A/q = 24), and, as a result, the single turn time must be 157 set with a high degree of precision. To determine this window, an off-line 158 scan is done of the opening time of the analyzer to find the region in which 159 the time of flight of the ions is undisturbed (see (20) for a full description 160 of the procedure). To double check this precision, an on-line test measuring 161 the mass of ${}^{24}Mg^+$ using ${}^{12}C_2^+$ as a calibrant at a similar number of turns 162 to the subsequent mass measurements while the opening time of the second 163 ion mirror was adjusted confirmed that we correctly set the single turn time. 164 This was confirmed due to the test measurements agreeing with literature 165 values within statistical uncertainty ($\delta m/m \approx 10^{-7}$). 166

¹⁶⁷ 4. Results and Discussion

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



Figure 3: A comparison of the different measurements for ²⁴Ne, ²⁵Ne and ²⁶Ne 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.

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

The ²⁵Ne ($T_{1/2} = 602$ ms) mass has been determined via Q values of 183 the reactions ${}^{26}Mg({}^{7}Li, {}^{8}B){}^{25}Ne$ at Berkeley (31) and ${}^{26}Mg({}^{13}C, {}^{14}O){}^{25}Ne$ at 184 Canberra (32), by direct mass spectrometry with MISTRAL (33; 34), and 185 by the CSR storage ring at Lanzhou (35). Although the reaction values 186 showed a roughly 2σ deviation, they were averaged in the AME, yielding 187 a mass compatible with our measurement, as is the one from the Lanzhou 188 storage ring. The MISTRAL value deviated by 3.4σ from the evaluated mass 189 so was rejected from the AME since the mass spectrum showed a double 190 peak. Coincidentally, the TITAN MR-TOF-MS was able to resolve a near 191 lying double peak caused by ${}^{12}C^{19}F_2^{2+}$ which could have been the cause of 192 the previously reported double peak. The TITAN MR-TOF-MS reached a 193 resolving power of $\approx 200,000$ in this experiment what the required resolving 194

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

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

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

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

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

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

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

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



Figure 4: The two-neutron separation energy (S_{2n}, Left) and empirical shell gap $(\Delta S_{2N}, \text{Right})$ 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).

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

229 5. Conclusion

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

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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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