Refining the nuclear mass surface with the mass of ¹⁰³Sn

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Mass measurements with the ISOLTRAP mass spectrometer at CERN-ISOLDE improve mass uncertainties of neutron-deficient tin isotopes towards doubly-magic ¹⁰⁰Sn. The mass uncertainty of 103 Sn was reduced by a factor of 4, and the new value for the mass excess of -67104(18) keV is compared with nuclear *ab initio* and density functional theory calculations. Based on these results and local trends in the mass surface, the masses of 101,103 Sn, as determined through their $Q_{\rm EC}$ values, were found to be inconsistent with the new results. From our measurement for ¹⁰³Sn, we extrapolate the mass excess of ¹⁰¹Sn to $-60\,005(300)$ keV, which is significantly more bound than previously suggested. By correcting the mass values for ^{101,103}Sn, we also adjust the values of ¹⁰⁴Sb, ^{105,107}Te, ¹⁰⁸I, ^{109,111}Xe, and ¹¹²Cs near the proton drip line which are connected through their α and proton Q-values. The results show an overall smoothening of the mass surface, suggesting the absence of deformation energy above the N = 50 shell closure.

INTRODUCTION I.

The heaviest self-conjugate (N = Z) nucleus bound against proton emission, ¹⁰⁰Sn, is one of the most interesting isotopes for nuclear research [1]. Its experimental investigation is important for testing our understanding of nuclear forces, especially since it is theorized to be a doubly magic nucleus with N = Z = 50 nucleons. First experimentally observed in the 1990s at GANIL, France [2], and at GSI, Germany [3], it has the smallest $\log(ft)$ value of any superallowed Gamow-Teller β -decaying nuclei [4], and it marks the endpoint of a secluded island of alpha-emitters [5, 6] and of the astrophysical rp-process [7–9].

Due to its small production cross-section through nuclear reactions [10], experimental knowledge on 100 Sn and its direct neighbors is, however, still sparse. Many investigations in this neutron-deficient area of the nuclear chart have been performed at facilities such as GSI, Germany, and RIKEN, Japan, using heavy primary beams such as ¹¹²In or 124 Xe to produce rare isotopes through fragmentation processes [4, 11–14] or fusion-evaporation reactions using the IGISOL technique [15, 16] in Jyvaskyla, Finland. Using the Isotope Separator Online (ISOL) technique with 1.4 GeV protons accelerated onto a thick target at CERN-ISOLDE in Switzerland [17], masses of indium isotopes (Z = 49) and their long-lived excited states were measured including ⁹⁹In, one proton below ¹⁰⁰Sn [18, 19], while laser spectroscopy has been performed on tin isotopes down to ¹⁰⁴Sn [20] and Coulomb excitation down to ¹⁰⁶Sn [21].

High-precision Penning-trap mass measurements on neutron-deficient tin isotopes were performed at IGISOL and GSI, reaching down to ¹⁰⁴Sn [8, 22]. More recently, the CSRe storage ring in Lanzhou, China, allowed the direct measurement of ¹⁰³Sn for the first time [23] followed by a Penning-trap measurement at FRIB, USA [24], and the TITAN ToF mass spectrometer at TRIUMF, Canada, measured down to ¹⁰⁴Sn [25].

Investigations of local trends of the mass surface surrounding ¹⁰⁰Sn have shown several inconsistencies when extracting masses from $Q_{\rm EC}$ -value measurements combined with masses of their daughter nuclei [18]. The mass for

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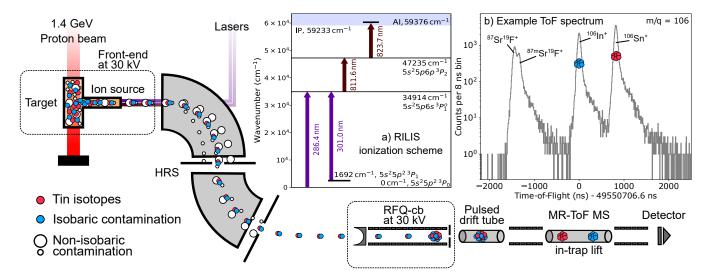


FIG. 1. Schematic of the apparatus. Protons with 1.4 GeV produce radioisotopes through nuclear reactions in the target material, where tin isotopes are vaporized and then laser-ionized through resonance laser ionization (laser scheme in insert a). Here, IP stands for the ionization potential of tin, and AI denotes the energy for auto-ionizing states. Tin isotopes (red circles) and parasitically ionized contamination (blue and white circles) are extracted at 30 keV and subsequently mass separated using two electromagnets (High-Resolution Separator, HRS). Ions with the selected mass-over-charge ratio m/q are collected in an ion cooler and buncher (RFQ-cb). The beam is then released in bunches, reduced in energy through the pulsed-drift tube to 3.2 keV, and captured in a time-of-flight spectrometer (MR-ToF MS) at 2 keV. After about 50 ms, the ions are released, and their flight time to an ion detector is measured. An example ToF spectrum for m/q = 106 showing strontium-fluoride, indium, and tin is depicted in insert b).

¹⁰⁰Sn itself is known only through two inconsistent $Q_{\rm EC}$ -value measurements [4, 12] in combination with the mass of ¹⁰⁰In [18]. Furthermore, the mass of ¹⁰³Sn, similarly extracted through a $Q_{\rm EC}$ -value, was not included in the latest Atomic Mass Evaluation (AME2020 [26]) due to its conflicting impact on the mass surface and a value derived from systematic trends was recommended instead.

In the present work, we report precision mass measurements of ¹⁰³⁻¹⁰⁶Sn performed by multi-reflection time-of-flight mass spectrometry at CERN-ISOLDE. We compare the results with nuclear *ab initio* [27, 28] and density functional theory calculations [29] and suggest several adjustments of neighboring masses based on this comparison and local binding energy trends in the region.

II. EXPERIMENT AND DATA ANALYSIS

A. Radioactive tin production at ISOLDE

Neutron-deficient radioactive tin isotopes were produced at CERN-ISOLDE [17] through proton-induced spallation of 139 La nuclei,

139
La $(p, 8p(19+x)n)^{113-x}$ Sn, (1)

driven by a 1.4 GeV proton beam delivered by the CERN Proton Synchrotron Booster (PSB) accelerator. Figure 1 shows a schematic of the apparatus for the production, separation, and mass measurement of the radioactive isotopes.

The target material in the form of lanthanum carbide with a density of 5 g cm^{-2} (ISOLDE target unit number #774) was bombarded by 1 to 2 µA of protons on average. Each PSB cycle, with a maximum repetition rate of one pulse every 1.2 s, contained between $1-3\times10^{13}$ protons per cycle. Reaction products diffused through and effused out of the porous target material into a small tantalum tube, connecting the target container to the hot-cavity resonance ionization laser ion source (RILIS) [30]. A three-step resonance ionization scheme was used to selectively ionize tin isotopes, shown in insert a) in the center of Fig. 1.

Different low-lying electronic states in the tin atom were populated as the radioactive isotopes were produced in the target container heated up to 1900 °C. To maximize ionization efficiency, two first excitation steps were used in parallel, starting from the $5s^25p^2 \ ^3P_0$ ground state and the first thermally populated state $5s^25p^2 \ ^3P_1$. Both transitions led to the $5s^25p6s \ ^3P_1$ state at $34\,914.28\,\mathrm{cm}^{-1}$, and the light used to excite them was generated via frequency-tripling

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a titanium:sapphire (Ti:Sa) laser to produce 286.4 nm and frequency-doubling a dye laser to produce 301.0 nm. The thermal population of the two levels strongly depends on the temperature of the ion source but is almost equal at 1900 °C. During the experiment, an ionization-efficiency enhancement of roughly 70% could be observed when using the transition from the thermally populated state in addition to the excitation of the ground state. A fundamental Ti:Sa laser drove the second step at 811.6 nm, exciting the $5s^25p6s \ {}^3P_1^o \rightarrow 5s^25p6p \ {}^3P_2$ transition. An autoionizing transition at 823.7 nm, driven by a Ti:Sa laser, was used as the final step.

The ionized tin isotopes were then extracted from the target and ion source front-end and accelerated as a beam of singly-charged radioactive ions at 30 keV. Tin ions of interest were separated from the extracted ion beam based on their mass-over-charge ratio m/q using the High Resolution Separator (HRS), which consists of two successive electromagnets depicted in dark grey in Fig. 1. The tin ions were finally delivered to the ISOLTRAP mass spectrometer [31], shown at the bottom of Fig. 1.

B. Mass measurement method

At the mass spectrometer, the ion beam was firstly decelerated and captured in a linear radio-frequency quadrupole cooler/buncher (RFQ-cb) [32], using helium buffer-gas cooling to improve beam emittance and to create a bunched beam with temporal bunch sizes in the order of several tens of nanoseconds. After a cooling time of 10 ms, the ion bunch was released from the trap and re-accelerated to 30 keV. To capture the ion bunch in the multi-reflection time-of-flight mass spectrometer (MR-ToF MS) [33], the ion kinetic energy was first reduced from 30 keV to 3.2 keV using a pulsed drift tube between the RFQ-cb and the MR-ToF MS, and once more from 3.2 keV to 2 keV using a pulsed drift tube in-between the two electrostatic mirrors of the mass spectrometer. Trapped inside the device, the ion bunch was reflected between the mirrors for up to 2000 turns before being ejected using the same pulsed drift tube, the so-called in-trap lift [34]. The ion ToF was then measured using a MagneTOFTM Mini 14924 electron multiplier detector downstream of the device.

The atomic mass m of the ions can be extracted using the measured time of flight t with respect to two reference masses m_1 and m_2 and their ToFs t_1 and t_2 as:

$$\sqrt{m} = C_{\rm ToF} \Delta_{\rm Ref} + \Sigma_{\rm Ref}/2,\tag{2}$$

where $\Delta_{\text{Ref}} = \sqrt{m_1} - \sqrt{m_2}$, $\Sigma_{\text{Ref}} = \sqrt{m_1} + \sqrt{m_2}$ and $C_{\text{ToF}} = (2t - t_1 - t_2)/[2(t_1 - t_2)]$ [35]. As reference ions, the isobaric indium isotope from the same spectrum and ⁸⁷Rb⁺ from interleaved reference measurements provided by a surface-ionization source at ISOLTRAP have been used. The mass-resolving power of the device is defined as

$$R = \frac{m}{\Delta m} = \frac{t}{2\Delta t_{\rm FWHM}},\tag{3}$$

where two different components in the ToF spectrum are considered fully resolved when their separation is at least twice the full-width-half-maximum (FWHM) of their ToF distribution.

The MR-ToF MS was operated in near-isochronous mode, in which faster ions penetrate the reflecting mirror potential deeper than slower ions, leading to longer turn-around times, thus compensating for their higher velocities. In such a way, the time focus was retained for long storage times, i.e. up to 50 ms, hence improving resolving power and resolution. This mode of operation was achieved by precisely tuning the ion kinetic energy inside the device by means of the in-trap lift voltage during pulsing. An example ToF spectrum for m/q = 106 is depicted in insert b) of Figure 1.

C. Target and ion source performance

Using the MR-ToF MS, the yield of the extracted neutron-deficient tin isotopes has been tracked over the period of the experiment using 106 Sn as a reference to monitor possible target micro-structure degradation and stability of the RILIS. Furthermore, 106 Sn⁺ served as a test case to optimize target and ion source heating parameters with respect to extraction efficiency and beam purity. Figure 2 (left) shows the normalized yield of the extracted 106 Sn⁺ versus time, together with target and ion source heating values. Yields in the order of 10^5 to 10^6 ions/s could be achieved throughout most of the experiment. However, towards the end, yields dropped on average by almost one order of magnitude.

At the beginning of the experiment, target and ion source heating were optimized based on the ¹⁰⁶Sn⁺ signal, shown in Fig. 2 (right). Lower ion source temperatures strongly suppressed isobaric contamination, such as indium and strontium monofluoride, which are parasitically surface-ionized in the hot environment of the ion source and

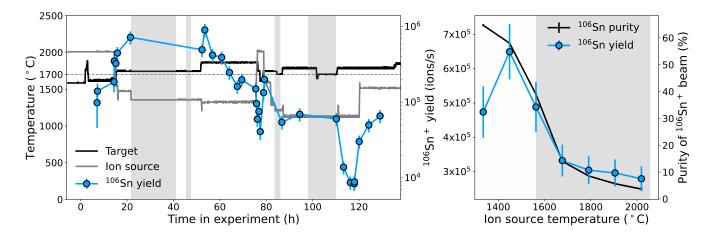


FIG. 2. Left: Normalized yields (in ions per second normalized to 1 μ A of protons on target) of the extracted ¹⁰⁶Sn⁺ beam as measured by ISOLTRAP throughout the experiment with respect to target and ion source temperatures. The temperatures were calibrated using an optical pyrometer for a range of applied heating currents and powers, with measurement uncertainties of about 50 °C (not shown). Shaded areas indicate where protons from the PSB were not available. The grey dashed line indicates the temperature at which the target material starts to undergo sintering. Right: Normalized yield and beam purity (percentage of tin ions in the MR-ToF spectrum) of the extracted ¹⁰⁶Sn⁺ beam versus ion-source temperature. The target temperature in the shaded area was 1650(50) °C, in the nonshaded area 1730(50) °C.

transfer line. The highest yields and lowest contamination levels were observed for increased target heating and lower ion source temperatures. High target temperatures facilitate the diffusion and effusion of reaction products through the target material and into the ion source. In contrast, a lower ion source temperature reduces the degree of parasitic surface ionization, improving the beam purity.

Besides the decrease in positive ion density, the thermal emission of electrons from the walls of the ion source decreases with lower temperature, while a surplus of negative charges is detrimental for ion confinement [36, 37]. Additionally, a lower heating current implies a smaller ohmic voltage gradient along the ion source, which leads to a slower drift of ions towards the extraction electrode. The increase in yield at lower ion source temperatures in Fig. 2 (right) is most likely due to a slightly lower target temperature $(1650(50) \,^{\circ}\text{C})$ as compared to the higher temperatures in the shaded areas of the figure $(1730(50) \,^{\circ}\text{C})$.

Due to the short half-lives of the tin isotopes of interest, the target was operated at or above 1700(50) °C to promote a quick diffusion and effusion from the target material. However, a drop in yield over time was observed, which can be attributed to the operation of the target container at temperatures at which lanthanum carbide undergoes a substantial degree of sintering, which leads to a decrease in the target material's open porosity [38] and results in a reduction of the effusion rate of the radioactive isotopes. Furthermore, the sublimation temperature of pure LaC₂ in vacuum is near 1700 °C, at which the target material starts to volatilize, further worsening the target's release properties. Additionally, the energy deposited by the pulsed proton beam leads to mechanical stress and a local hotspot, accelerating the deterioration of the target microstructure.

Figure 3 shows the best yields for observed neutron-deficient tin isotopes. These are compared with previously reported values from the ISOLDE yield database [39, 40], to in-target production simulations (available in the same database) [41–43], and to empirical in-target production cross-section calculations [44]. On average, the yields were significantly lower than previously reported best yields from the same target and ion source combination at ISOLDE (compare red and blue diamonds) [20, 39]. The measured yield for 105 Sn⁺ is disproportionately lower than the trend would suggest, most likely caused by a low ion source temperature of 1150(50) °C used during that specific measurement, which was also observed to have a negative impact on the extracted yield of 106 Sn (see Fig. 2 (right)).

The trends and values of the different production simulations for the tin isotopes closer to stability match the measurements within one order of magnitude on average, as most of the produced isotopes in the target can be extracted and ionized due to their long half-lives. For isotopes with half-lives well below 1×10^4 s, the trend of the extracted ions deviates significantly from the calculated in-target production cross-section, indicating large losses during the extraction process due to target-release characteristics, ionization efficiency, and radioactive decay. These losses become significant below half-lives of 100 s, where the extracted yields differ by more than two orders of magnitude, highlighting the challenges of diffusion of isotopes inside the target material in the ISOL production method [45].

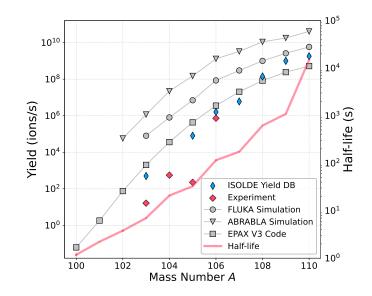


FIG. 3. Best yields extracted from the target measured with the MR-ToF MS (red) and normalized to a proton current of 1 µA. In comparison, previously measured yields of extracted beams reported in the ISOLDE yield database (blue) and in-target production simulation codes FLUKA [41, 42] and ABRABLA [43], as well as the empirical fragmentation production cross section code EPAX-V3 [44].

D. ToF spectra and fitting

Figure 4 shows ToF data of the four different mass measurements presented in this study. In all spectra, the surface-ionized strontium-fluoride and indium contaminants are present. While for m/q = 106, this contamination could be reduced to less than 40% (shown in Fig. 2 (right)), for smaller m/q values, the contamination gradually increased to >99.9% of the isobaric beam.

To calculate the atomic masses as shown in Eq. (2), the Tof was extracted by fitting a multi-component exponentially modified Gaussian distribution ("hyper-EMG") [46] to the data. Fits with different components were tested to determine the necessary number of components for the fit function, and the resulting reduced chi-squared $\chi^2_{\rm red}$ was observed. One negative component and three positive components resulted in $\chi^2_{\rm red} < 10$, sufficiently representing the data without leading to parameter overfitting. The final fit was performed using unbinned maximum likelihood estimation, implemented in CERN's RooFit MINUIT minimizer [47]. With 2000 revolutions resulting in storage times of up to 50 ms in the MR-ToF MS, mass resolving powers R of up to 300 000 were achieved during the experimental campaign, which proved sufficient to resolve the isomeric states in the odd-even indium isotopes and in ⁸⁷Sr (with excitation energies of about 650 keV and 390 keV, respectively).

Due to the intense contamination levels on m/q = 104 and m/q = 103, only the ToF range between indium and tin was fitted for all masses. This prevented systematic effects resulting from detector and data acquisition saturation around the arrival time of the SrF⁺ ions from being introduced into the shape of the fit function. For the fitted time range, the average count rate was well below five ions per experimental cycle, which prevented detector pileup (the single-ion detector response of the detector is about 1.5 ns long for a MagneTOF^M Mini 14924) nor to loss of signals due to data acquisition deadtime (<100 ps for FAST ComTec MCS6A). Furthermore, the ion load in the MR-ToF MS was kept well below 20 ions per cycle to avoid space-charge effects that can lead to systematic ToF shifts [48].

In Fig. 4a, an example of the spectra for m/q = 106 is shown, where one can see, from left to right, the contamination 87 Sr¹⁹F⁺ and 106 In⁺, as well as the isotope of interest 106 Sn⁺. Interestingly, as noticed during the systematic measurements of this mass-over-charge ratio throughout the experiment, the shoulder on the 87 Sr¹⁹F⁺ peak only appeared after several tens of hours into the experiment. The observed ToF matches that of a strontium-fluoride molecule containing the long-lived I = 1/2 isomeric state with an excitation energy of 389 keV, leading to a slightly longer ToF. The involved time scales of the isomer's appearance suggest that the strontium isomer is not directly produced through nuclear reactions but rather through the in-target beta-decay of the I = 1/2 ground state of 87 Y with a half-life of almost T = 80 h [49]. A typical measurement for the beam composition on this m/q value took five minutes. Figure 4a shows data for about 20 min of measurement time.

Figure 4b shows the data taken for m/q = 105, where ${}^{105}\text{Ag}^+$, ${}^{86}\text{Sr}{}^{19}\text{F}^+$, ${}^{105g,m}\text{In}^+$, and ${}^{105}\text{Sn}^+$ were present in the beam. In this spectrum, the I = 1/2 isomer of ${}^{105}\text{In}$ was observed with an abundance of about 1:80 with respect

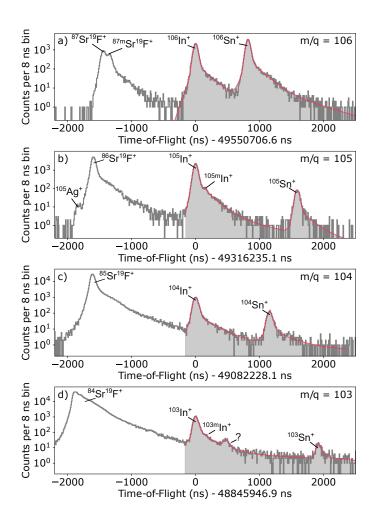


FIG. 4. Typical ToF spectra for the four tin isotopes investigated in the present study. The grey-shaded area indicates the fit range, which excludes the potentially saturated peaks at smaller ToFs. The red line shows the fit to the data. The horizontal axis is centered at the indium peak.

TABLE I. Mass measurement results in comparison with storage ring measurements from [23], MR-ToF MS measurements from [25], and the AME2020, which is based on the Penning trap measurements reported in [8]. Spin assignments J^{π} , half-lives, and reference masses (^AIn for mass m_1 , and ⁸⁷Rb for mass m_2) are taken from the AME2020. Values marked with # are extrapolated or assigned from systematics.

						Mass excess (keV)			
1	4	J^{π}	Half-life	Ref. ions	C_{ToF}		AME2020 [51]		TITAN [25]
1	103	$5/2^+ \#$	$7.0(2){ m s}$	$^{87}\text{Rb}^+, ^{103}\text{In}^+$	0.500484342(920)	-67104(18)	-67090(100)#	-67138(68)	/
		$0^+ \#$	$20.8(5){ m s}$	87 Rb ⁺ , 104 In ⁺	0.500275256(476)	-71629(10)	-71627(6)	/	-71601(50)
					0.500358319(329)			1	-73349(34)
1	106	$0^+ \#$	$1.92(8) \min$	$^{87}\text{Rb}^+, ^{106}\text{In}^+$	0.500175627(71)	-77346(14)	-77354(5)	/	-77327(37)

to the I = 9/2 ground state, and the spectrum was fitted with three individual peaks. The data for this spectrum corresponds to a measurement time of about 800 s.

For the ion beam with m/q = 104, ${}^{85}\text{Sr}^{19}\text{F}^+$, ${}^{104}\text{In}^+$, and ${}^{104}\text{Sn}^+$ were observed, and the spectrum was fitted with only two individual peaks. Interestingly, the I = 1/2 isomeric state in the ${}^{85}\text{Sr}$ containing molecule was not observed, despite the predominant beta-decay of ${}^{85}\text{Y}$ into the isomeric state [50]. The data within this plot was taken in about 1 h of measurement.

On m/q = 103, one can notice the increase of contamination in the beam: the ⁸⁴Sr¹⁹F⁺ peak on the left is saturating the discriminator-based data acquisition (note the double-peak structure and the pronounced tailing). Again, both isomer and ground state in ¹⁰³In⁺ were resolved. However, one additional peak appears between the two indium states and ¹⁰³Sn⁺ on the right-hand side of the spectrum, which is inconsistent with any isobaric mass or previously observed

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surface-ionized molecule. It can not be ruled out that this species was ejected from the MR-ToF MS on a different revolution number than the other isotopes, hence not having traveled the same distance. Such contamination can usually be investigated by varying the overall trapping time and observing the relative number of peaks in the spectra. This was not attempted due to the limited time available for these m/q settings. As mentioned in Section II C, due to the high operation temperature and proton current during long periods of time, which most likely led to target material sintering, yields for the more short-lived tin isotopes dropped quickly throughout the experiment. As a result, the ratio of 103 Sn⁺ versus contamination in the beam worsened throughout the experiment, preventing the measurement of the lighter neutron-deficient tin isotopes. Effectively, 2.4 h of data from the first day after the start of the experiment are included in the plot and the analysis of the final mass value.

III. RESULTS AND DISCUSSION

Table I shows the results for $^{103-106}$ Sn, together with available literature values on mass excesses and half-lives, as well as tentative spin assignments. The references for extracting the mass of the ion of interest were consistently taken from the isobaric indium ions and from 87 Rb⁺ provided by the ISOLTRAP offline ion source. All mass excess values are in very good agreement with the available literature data of the AME2020 [51], the storage ring measurements from Lanzhou [23], and the MR-ToF MS measurements from TITAN at TRIUMF [25]. The experimental uncertainty is between 10 keV and 20 keV, reaching a precision of about $\delta m/m \sim 2 \times 10^{-7}$. The reported mass on 103 Sn is a factor four more precise than the storage ring measurement (-67 138(68) keV) [23] and, in combination with the other mass measurements, allows evaluation of the mass surface in the neutron-deficient tin region. With this new measurement, we can deduce the $Q_{\rm EC}$ value for the decay of 103 Sn into 103 In to be 7528(20) keV, compared to 7640(70) keV reported in [52] which was extracted from total absorption spectroscopy and beta-delayed proton energy measurements.

To investigate the impact of the shift and increased precision of the 103 Sn mass measurement on the local mass surface, the two-neutron separation energy

$$S_{2n}(Z,N) = B(Z,N-2) - B(Z,N)$$
(4)

and the odd-even staggering estimator

$$\Delta_{3n} = 0.5 \cdot (-1)^N \cdot [B(Z, N-1) - 2 \cdot B(Z, N) + B(Z, N+1)],$$
(5)

are calculated, where $B(Z, N) = M_E(N, Z) - Z \cdot M_E(^1H) - N \cdot M_E(n)$ is the total nuclear binding energy which consists of the nuclear mass excess $M_E(N, Z)$ of the nucleus of interest, and the mass excesses of the hydrogen atom ¹H and the mass excess of the neutron. The S_{2n} filter is regularly used to evaluate shell closures and the sudden change of nuclear deformation. Typically, one analyzes the change of slope in S_{2n} values with the addition or removal of neutrons, especially at magic numbers. Complementarily, the Δ_{3n} filter quantifies the strength of the odd-even staggering along an isotopic chain.

The two mass filters are plotted in Fig. 5, comparing mass values extracted based on $Q_{\rm EC}$ values for $^{101-103}$ Sn (grey) and direct mass measurements for $^{103-108}$ Sn (red) with nuclear *ab initio* calculations from [18] and density functional theory (DFT) calculations described in Ref. [53] and references therein. In the upper panel, using the S_{2n} filter, one can observe a smooth evolution in the binding energy differences above the expected shell closure at N = 50. This smooth trend is irregular at N = 53 and at N = 55 when using masses for $^{101-103}$ Sn derived using $Q_{\rm EC}$ links. This irregularity is further highlighted by plotting the odd-even staggering estimator, shown in the lower panel of Fig. 5.

Extracting masses through β end-point energies requires complete knowledge of the feeding into excited states in the daughter nucleus to reliably fit the β -decay spectrum. High-resolution γ -spectroscopy of the decay is needed to account for all internal transitions and total absorption spectroscopy, as well as energy measurements of the delayed proton emission to draw a full picture of the decay. However, incomplete knowledge of the full decay chain caused by internal γ conversion or undetected weak decay branches may lead to significant systematic errors. In the 2020 Atomic Mass Evaluation of input data [26], the mass of ¹⁰¹Sn is extracted through a β -delayed proton decay channel via ¹⁰¹In into ¹⁰⁰Cd, with a dominating mass uncertainty of 300 keV assigned from a measurement of the proton emission energy and branching ratios, reported in a scientific report at GSI [54]. Similarily, the ground-state mass of ¹⁰²Sn is extracted from the ¹⁰²In ground-state mass using the $Q_{\rm EC}$ value measured in [55], incorporating the 100 keV measurement uncertainty. Additionally, ¹⁰³Sn was deemed a "seriously irregular mass" determined only from the ¹⁰³In ground-state mass through the $Q_{\rm EC}$ value measurement from Ref. [52], which led to its rejection from the AME2020 [26]. Finally, the precise mass measurement of ¹⁰⁰In from Ref. [18] highlighted another discrepancy, namely

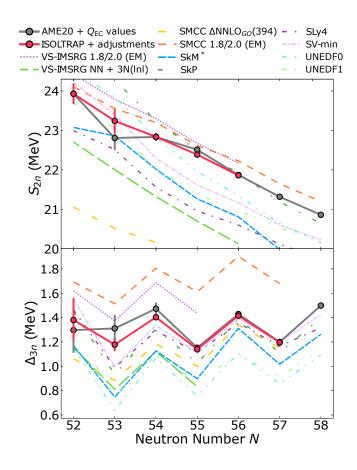


FIG. 5. Evaluated two-neutron separation energy (top) and odd-even staggering estimator (bottom) from masses reported in the AME2020 (grey), the ISOLTRAP measurement with adjustments (red), nuclear *ab initio* calculations from [18], and DFT calculations from [53]. The grey data points include the masses of $^{101-103}$ Sn calculated with the $Q_{\rm EC}$ value measurements from [52, 54, 55]. The red points include this work's mass measurement of 103 Sn and the extrapolated mass value for 101 Sn.

the competition between the results of the two direct beta-decay measurements of ¹⁰⁰Sn [4, 12], whose $Q_{\rm EC}$ values disagree on a 2σ uncertainty level.

Based on the present precise mass measurements, we reevaluate the mass values expected from systematic trends and from comparing them to theoretical calculations. The present ¹⁰³Sn mass excess of $-67\,104(18)\,\text{keV}$ makes this nucleus about 130 keV less bound compared to the value of $-66\,970(70)\,\text{keV}$ extracted from the Q_{EC} value. However, to fully remove the kink at N = 53, one must also adjust the mass of ¹⁰¹Sn, which contributes in equal parts to the S_{2n} value. By adding 300 keV to the mass excess of ¹⁰¹Sn, within two standard deviations of the reported betadelayed proton energy measurement [54], one obtains a smooth trend, as highlighted by the red data point in Fig. 5. We motivate this adjustment not only by comparing the local trends of the mass surface near the shell closure in N = 50 but also based on the predicted trends by the nuclear *ab initio* and DFT calculations. While not all methods, interactions, and functionals predict the correct magnitude of the S_{2n} value, all calculations predict a flat trend.

Contrary to extracting masses through beta-decay endpoint energies, nuclear α -decay is an intrinsically accurate method as the kinetic energies of the emitted alpha-particle are mono-energetic and relatively easy to measure with keV precision. A unique feature of the nuclear landscape is the small island of α -emitters with decay chains ending in neutron-deficient tin isotopes. Most of the mass data of those short-lived isotopes are indirectly known based on anchoring tin masses through successive α -decays by measuring the α -particles energies, see for example Ref. [5, 6]. By directly measuring ¹⁰¹⁻¹⁰³Sn, one can significantly reduce the mass uncertainties for antimony, tellurium, iodine, xenon, and cesium isotopes, which are otherwise very challenging for ion-trap-based atomic mass measurements. Table II shows mass excess values, which are based on our ¹⁰³Sn mass measurement combined with the measurement of Ref. [23] and the proposed ¹⁰¹Sn mass adjustment.

The masses from Tab. II in combination with the mass tables from the AME2020 can be used to plot the S_{2n} values for a large range of isotopes (Z = 41 - 56), shown in Fig. 6. On a more global picture, the trends for the S_{2n} values are extremely smooth above the N = 50 shell closure for nuclei below with Z < 50, where all necessary masses for

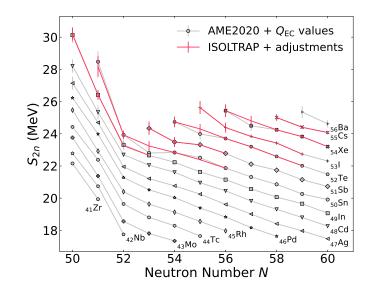


FIG. 6. Evaluated two-neutron separation energy from masses reported in the AME2020 including the masses of $^{101-103}$ Sn calculated from $Q_{\rm EC}$ value measurements [52, 54, 55] (grey) or readjusted as shown in Tab. II following the ISOLTRAP mass measurements from this work (red). Plotted improvements on indium mass uncertainties are taken from [18, 19].

the evaluation are known to sufficient precision. This smoothness has now been recovered for the tin chain. Above Z = 50, one notices significant changes at N = 55 in the tellurium chain and at N = 57 in the xenon chain, leading to overall smoother trends as well.

IV. CONCLUSION

Multi-Reflection ToF Mass Spectrometry was performed on $^{103-106}$ Sn isotopes produced at the radioactive ion beam facility at CERN-ISOLDE. Radioactive tin isotopes were produced through nuclear reactions of high-energy protons impinging on a LaC_x target and were ionized through a three-step laser ionization scheme. The target and ion source performance was tracked based on yields of 106 Sn⁺, which dropped with time, likely because of sintering of the target material. This study offers a detailed basis for future developments of neutron-deficient tin beams at ISOL facilities and strongly motivates further improvements of ISOL target materials and their degradation over time. The highest yields of extracted $^{103-106}$ Sn⁺ ions were compared to in-target production simulations and empirical cross-section calculations, showing slightly lower values than previously achieved during other experiments at ISOLDE.

All cases suffered from surface-ionized contamination and target degradation, preventing mass measurements of lighter tin isotopes despite sufficient production rates initially estimated from simulations. Atomic masses for $^{103-106}$ Sn were obtained, with uncertainties ranging between 10 keV and 20 keV. The mass uncertainty of 103 Sn was improved by roughly a factor of four. The measured masses were compared with nuclear *ab initio* and DFT calculations to

TABLE II. Mass excess values derived from the extrapolated mass of 101 Sn and present measurements of 103 Sn combined with Ref. [23]. The re-evaluated mass excess values are compared to those reported in the AME2020 [51]. Values marked with # are extrapolated from the mass surface, and values marked with * are based on the present adjustment of 101 Sn.

	Mass excess (keV)	AME2020 (keV)
101 Sn	-60005(300)*	-60305(300)#
$^{103}\mathrm{Sn}$	-67105(16)	-67092(100)#
$^{104}\mathrm{Sb}$	-59307(22)	-59340(100)#
$^{105}\mathrm{Te}$	-52510(300)*	-52810(300)#
$^{107}\mathrm{Te}$	-60670(17)	-60660(100)#
^{108}I	-52784(21)	-52770(100)#
$^{109}\mathrm{Xe}$	-45870(300)*	-46170(300)#
111 Xe	-54530(60)	-54520(120)#
$^{112}\mathrm{Cs}$	-46430(60)	-46415(120)#

assess the local smoothness of the mass surface near ¹⁰⁰Sn. It was shown that the extracted masses for ^{101,103}Sn from $Q_{\rm EC}$ value measurements are very likely inaccurate and lead to discrepant trends. Using the more precise mass value of ¹⁰³Sn in combination with a mass extrapolation for ¹⁰¹Sn, this irregularity was removed, recovering smooth trends not only in the tin chain but also in the tellurium and xenon chains.

The mass uncertainty of five other nuclides near the proton drip line, linked to the tin masses through α -decays, was thus improved. This result highlights the necessity for precision mass spectrometry of short-lived radioactive nuclei, since inaccurate beta-decay measurements with significant measurement uncertainties — in the order of a few hundreds of keV — can produce misleading trends of the mass filters.

In the course of submission of this article, we were made aware of a recent preprint [24] reporting a Penning-trap mass measurement value for ¹⁰³Sn with a measured mass excess of -67125.9(37) keV. The authors of Ref. [24] draw a similar conclusion considering the smoothness of the mass surface in the tin isotopic chain near N = 53.

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