

# Precision Mass Measurements Reveal Low Neutron Pairing in Tin beyond $N = 82$ and Its Impact on Stellar Nucleosynthesis

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We present a study on neutron-rich tin ( $Z = 50$ ) isotopes beyond the doubly closed shell of  $N = 82$  through high-precision mass measurements, including the first-ever measurements of the masses of  $^{136}\text{Sn}$ ,  $^{137}\text{Sn}$ , and  $^{138}\text{Sn}$  isotopes. These measurements enhance our understanding of the nuclear structure and astrophysical nucleosynthesis in this previously unexplored region. The new mass data are used for evaluation of the final abundances of mass numbers  $A = 135$  and  $137$  in  $r$ -process network calculations. Our findings reveal a notable change in the empirical pairing gap for tin isotopes beyond the  $N = 82$  closed shell and a shift in the two-neutron-separation energy slope compared to heavier elements above the shell closure. A new set of *ab initio* calculations effectively describes these observed trends.

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Studying exotic nuclei with extreme neutron-to-proton ratios is crucial for understanding fundamental nuclear properties and the formation of heavy elements in the

Universe [1–3]. Atomic mass measurements are essential for probing nuclear structure and are a necessary input for nucleosynthesis calculations.

The 2017 observation of a kilonova (GW170817) [4,5] from electromagnetic radiation, combined with gravitational waves from the merging of two neutron stars [6], garnered significant scientific attention [7–9]. This event highlighted a fundamental question: how are heavy elements produced in astronomical environments [10]? The early blue emission and gradual shift to red in the light spectrum confirmed the formation of heavy nuclei via the

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rapid-neutron capture process ( $r$  process) [6]. However, the models describing the  $r$  process cannot precisely predict the abundances of heavy elements due to a limit of experimental knowledge of the heavy neutron-rich nuclei involved and the astrophysical conditions. These nuclides are only produced in astronomical events and have short lifetimes [11]. To address this, extensive collaborative efforts are underway at accelerator-based facilities worldwide to produce, identify, and measure the properties of these rare nuclides.

Isotopes with closed shells of protons or neutrons are frequently used as testing grounds for nuclear physics [12–17]. The tin isotopic chain with a magic proton number of  $Z = 50$  is the longest isotopic chain, including two closed neutron shells ( $N = 50, 82$ ). However, our understanding of nuclear properties around these closed shells remains limited, as producing these exotic nuclei is at or beyond the capabilities of current facilities. The neutron-rich side of the tin isotopic chain is particularly intriguing because the expected  $r$ -process path overlaps with unknown nuclear structure properties for very neutron-rich nuclei [18–20].

A proton-to-neutron ( $p$ -to- $n$ ) converter target was recently implemented at ISAC, TRIUMF, aiming to produce cleaner and higher yields of fission fragments. This development builds on existing technology [21], but it is the first realization at TRIUMF [22]. Here, we present the first physics results, demonstrating the significant potential of this development. A proton beam of up to 50  $\mu\text{A}$  and an energy of 480 MeV was directed into the  $p$ -to- $n$  target to produce rare nuclei via neutron-induced fission on natural uranium. A three-step resonant laser-ionization scheme (with wavelengths 301.001, 811.62, and 823.67 nm in vacuum) was used for the ionization of the tin isotopes [23]. The ions were transported to the TITAN experiment [24], where the multiple reflection time-of-flight mass spectrometer (MR-TOF-MS) [25–27] was used for the yield measurements as well as high-precision mass measurements [28–31]. The high sensitivity of the MR-TOF-MS allowed us to perform mass measurements with production yields as low as 0.08 particles per second (pps) for  $^{138}\text{Sn}$  ( $1.6 \times 10^{-3}$  pps/ $\mu\text{A}$  proton beam). This corresponds to a detection rate of 30 per hour at the MR-TOF-MS detector with a 10% transport and detection efficiency (Fig. 1). Additionally, the MR-TOF-MS can serve as an isobar separator using the mass selective Re-trapping method [32,33]. This method was utilized during the experiment to suppress the isobaric contamination (Cs, Ba). Ions of interest traveled in the analyzer of the MR-TOF-MS around 700 isochronous turns, achieving a mass resolving power of around 450 000 in just 33 ms measurement cycle time. The atomic masses of tin isotopes were determined using well-known isobaric species as mass calibrants. Identification of tin isotopes was further confirmed by blocking the selective lasers for resonance ionization, which resulted in the

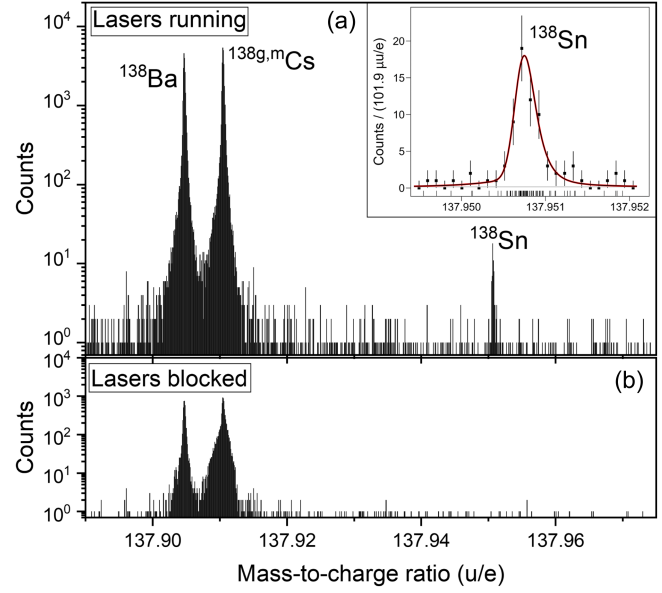


FIG. 1. The measured mass spectrum of  $A = 138$  with and without laser ionization in (a) and (b), respectively. The inset in (a) shows an enlargement on  $^{138}\text{Sn}$  fitted with a hyper exponentially modified Gaussian [35] (red line). Data were accumulated for about 133 and 33 min with and without laser ionization, respectively.

complete disappearance of the tin peaks. More details on the data analysis methods can be found in Refs. [34,35].

Table I summarizes the mass excess (ME) values of tin isotopes measured by TITAN and compares them with the literature mass values from AME2020 [36]. The AME2020 presents the weighted average of the two previous mass measurements in Refs. [18,37]. The ME values measured in this study are in very good agreement with [18,37] and include the first-time mass measurements of heavy Sn isotopes with the mass numbers  $A = 136, 137$ , and 138.

TABLE I. The measured mass excess (ME) value and one standard deviation uncertainty ( $\sigma$ ) of the neutron-rich tin isotopes compared to the literature values (AME2020) [36]. The masses of tin isotopes with atomic mass numbers  $A = 136, 137$ , and 138 were measured for the first time. The hashtag shows the AME2020 theoretical extrapolation values.

| Isotope           | Calibrant          | ME <sub>TITAN</sub><br>(keV) | ME <sub>AME20</sub><br>(keV) | Difference<br>(keV) |
|-------------------|--------------------|------------------------------|------------------------------|---------------------|
| $^{131}\text{Sn}$ | $^{131}\text{Cs}$  | $-77\,268 \pm 22$            | $-77\,265 \pm 4$             | $-3 \pm 23$         |
| $^{132}\text{Sn}$ | $^{132}\text{Cs}$  | $-76\,539 \pm 11$            | $-76\,547 \pm 2$             | $7 \pm 11$          |
| $^{133}\text{Sn}$ | $^{133}\text{Cs}$  | $-70\,870 \pm 24$            | $-70\,874 \pm 2$             | $4 \pm 24$          |
| $^{134}\text{Sn}$ | $^{134}\text{Ba}$  | $-66\,445 \pm 26$            | $-66\,434 \pm 3$             | $-11 \pm 27$        |
| $^{135}\text{Sn}$ | $^{135m}\text{Cs}$ | $-60\,647 \pm 15$            | $-60\,632 \pm 3$             | $-15 \pm 15$        |
| $^{136}\text{Sn}$ | $^{136}\text{Ba}$  | $-56\,074 \pm 29$            | $-56\,170^{\#} \pm 200^{\#}$ | 96                  |
| $^{137}\text{Sn}$ | $^{137}\text{Cs}$  | $-50\,110 \pm 10$            | $-50\,150^{\#} \pm 300^{\#}$ | 40                  |
| $^{138}\text{Sn}$ | $^{138}\text{Ba}$  | $-45\,442 \pm 21$            | $-45\,510^{\#} \pm 400^{\#}$ | 68                  |

In the following discussion, we examine the new masses by analyzing the empirical pairing gap and its impact on the nuclear structure beyond the neutron shell closure. The empirical pairing gap is derived from the measured atomic masses ( $M$ ) of three isotopes:

$$\Delta_C^{(3)}(N) = [2M(N-1, Z) - M(N, Z) - M(N-2, Z)] \times c^2/2, \quad (1)$$

for the even-even nuclei [38]. This observable is sensitive to the change of nuclear structure along the isotopic chain as the number of valence neutrons changes. Figure 2 shows the  $\Delta_C^{(3)}(N)$  along the Sn isotopic chain ( $Z = 50$ ). Here, the experimental values show a drop after the shell closure of  $N = 82$ . One possible explanation given in Ref. [37] is the variation of the empirical pairing gaps due to the change in level density and spin orbitals before and after the neutron shell closure. The new TITAN mass measurements confirm that the trend of the empirical pairing gap remains significantly lower after the  $N = 82$  shell closure. A weaker reduction of pairing gap is shown in Ref. [37] for heavier elements  $Z > 50$ . For  $Z < 50$ , the recent mass measurements of Cd isotopes around the  $N = 82$  [39] exactly reproduce the reduction strength of pairing gap as in the Sn isotopic chain after  $N = 82$ .

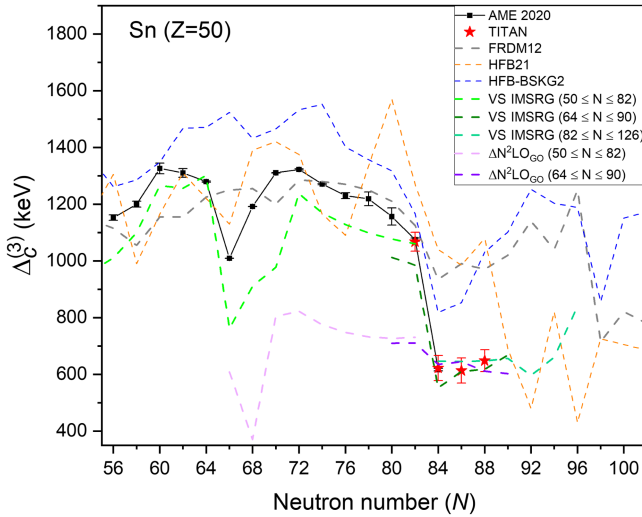


FIG. 2. The empirical pairing gap  $\Delta_C^{(3)}(N)$  is plotted against the neutron number ( $N$ ). The black data points are calculated by using the atomic mass values reported in the literature (AME2020) [36], while the red data points represent the new mass measurements. These are compared to empirical pairing gap obtained from common theoretical mass models: FRDM12 (gray dashed line) [40], HFB21 (orange dashed line) [41], and HFB-BSKG2 (blue dashed line) [42]. Additionally, new sets of *ab initio* calculations are also performed in this work, based on the different valence nucleons in the neutron shell, and are shown as green and purple dashed lines.

The neutron pairing gap across other shell closures, e.g., for the lighter shells of Ca, Ni, and Zr or heavy Pb, are behaving differently; this can be understood due to different orbitals and level densities involved.

It appears that the theoretical mass models (FRDM12, HFB21, and HFB-BSKG2) are not capable of predicting the empirical pairing gap beyond  $N = 82$  (gray, orange, and blue dashed lines in Fig. 2) within their expected uncertainties (a few 100 s keV) [42,43]. The HFB21 model suggests a smooth reduction of the empirical pairing gap beyond the shell closure for heavy isotopes after  $N = 82$ . The more sophisticated calculations including SLy4 interaction and mean-field theories have qualitatively addressed the pairing gap reduction beyond  $N = 82$  [37,44]. A calculation based on the  $\Delta N^2LO_{GO}$  interaction does not capture the change at  $N = 82$ .

A precise description of  $\Delta_C^{(3)}(N)$  is only achieved, without tuning the pairing interactions, in the valence-space formulation of the *ab initio* in-medium similarity renormalization group (VS IMSRG) model performed in this work. The calculations reproduce the  $\Delta_C^{(3)}(N)$  minima at  $N = 66$  and also the behavior beyond  $N = 82$ , in agreement with the new results reported here. Here we perform calculations with the VS IMSRG [45], utilizing different valence spaces, depending on which region of the isotopic chain is being studied. The VS IMSRG calculations are performed with the EM 1.8/2.0 interaction [46,47], which can reproduce the experimental ground-state energies and other structure properties throughout the medium-mass region [48] to the tin region [49–52]. The calculations start from the 15-major-shell spherical harmonic-oscillator (HO) basis, where for the three-nucleon ( $3N$ ) forces, an additional  $E_{3\max}$  truncation defined as the sum of the three-body HO quanta needs to be introduced. Here we use sufficiently large  $E_{3\max} = 24$  [53] which has been shown to converge ground-state energies even to the  $^{208}\text{Pb}$  region [54]. Finally, to best capture  $3N$  interactions between valence nucleons, the ensemble normal ordering technique [55] is used to construct the reference state for each individual isotope. We evolve the Hamiltonian with the IMSRG(2) where many-body operators arising in the flow are truncated at the two-body level, decoupling the targeted valence-space Hamiltonian from the full-space Hamiltonian. For  $N \leq 82$  ( $N \geq 82$ ) systems, the neutron  $\{2s_{1/2}, 1d_{3/2}, 1d_{5/2}, 0g_{7/2}, 0h_{11/2}\}$  ( $\{2p_{1/2}, 2p_{3/2}, 1f_{5/2}, 1f_{7/2}, 0h_{9/2}, 0i_{13/2}\}$ ) orbitals are taken as the valence space. In addition, to investigate a valence-space artifact at  $N = 82$ , we also use neutron  $\{2s_{1/2}, 1d_{3/2}, 0h_{11/2}, 1f_{7/2}\}$  valence space near  $N = 82$  using the multishell formulation of the VS IMSRG, described in Ref. [56]. For the VS IMSRG calculations, we used the NuHamil code [57] to generate the  $NN + 3N$  matrix elements, IMSRG++ code [58] for the VS IMSRG decoupling, and KSHELL code [59] for the valence-space diagonalizations.

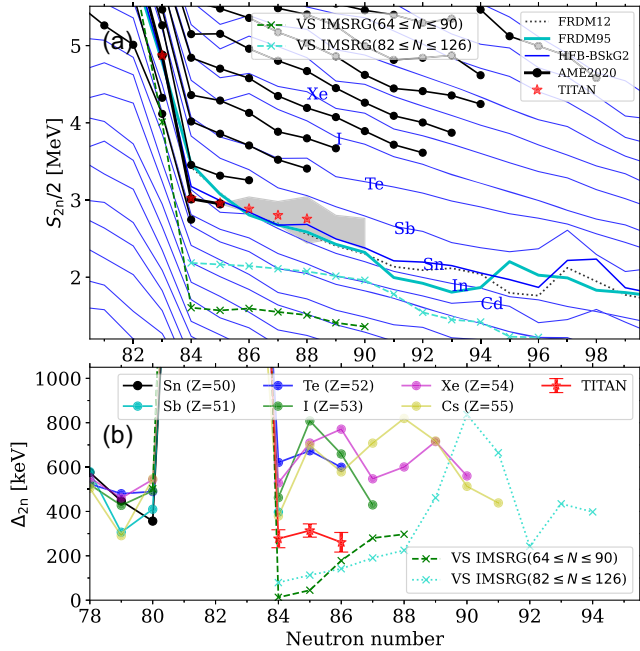


FIG. 3. (a) The two-neutron separation energy  $S_{2n}$  for neutron-rich isotopes around the  $N = 82$  is depicted. The blue lines represent the values calculated for the HFB-BSkG2 mass model [42] for all elements, while the black lines with filled circles show the experimental values from AME2020. For  $Z = 50$ , the  $S_{2n}$  values from the FRDM95, the FRDM12 mass models [40], and the current experimental values are also included. The error bars of the TITAN values are too small to be seen. The gray-shaded area indicates the estimated range from the AME2020 extrapolation. (b) The two-neutron shell gap from AME2020 is shown for  $Z = 50$ – $55$ . The results from this study and from the *ab initio* calculations (indicated by dashed and dotted lines in both panels; see text) are presented for  $Z = 50$ .

Another important observable for probing nuclear structure, which is also crucial for nuclear astrophysical calculations, is the two-neutron-separation energy,  $S_{2n} = [M(Z, A - 2) - M(Z, A) + 2M_n] \times c^2$ , where  $M(Z, A)$  and  $M(Z, A - 2)$  are the mass of bound nuclei which differ in two neutrons,  $M_n$  is the rest mass of a free neutron, and  $c$  is the speed of light in vacuum. Figure 3(a) illustrates the trends of  $S_{2n}$  around  $Z = 50$ , showing a sharp drop at  $N = 82$  due to the shell closure. This drop is responsible for the second abundance peak in *r*-process nucleosynthesis. When an equilibrium between  $(n, \gamma)$  and  $(\gamma, n)$  reactions is established, the *r* process tends to populate isotopes along a constant  $S_{2n}$  value, determined by the environment's temperature and density [60–63]. In this scenario, sudden changes of slope of  $S_{2n}$  values in isotopic chains result in troughs in the abundance pattern [64]. Figure 3(a) indicates that all theoretical mass models predict relatively large gaps between  $Z = 50$  and  $Z = 51$  for  $N > 90$ . This leads to the trough in the abundance pattern between  $A = 140$  and  $A = 144$ , as shown in our calculations below (Fig. 4). For  $Z = 50$ , predictions from the recent FRDM12

and HFB-BSkG2 models are very similar, but the older FRDM95 model shows a particularly pronounced gap in  $S_{2n}$ . Our experimental results have significantly extended the known  $S_{2n}$  region (see the red stars) in Fig. 3. These results also allow us to exclude any unexpected strong variations in  $S_{2n}$  up to  $N = 88$  due to the observed smooth trend.

Moreover, the slope of the  $S_{2n}$  beyond the shell closure for  $Z = 50$  is significantly less steep compared to that of heavier elements with  $Z > 50$  [Fig. 3(a)]. The slope of  $S_{2n}$ , referred to as the two-neutron shell gap [ $\Delta_{2n} = S_{2n}(Z, N) - S_{2n}(Z, N + 2)$ ], is shown in Fig. 3(b). The tin isotopes ( $Z = 50$ ) exhibit the lowest values after the shell closure, in comparison to the elements with  $Z > 50$ . Additionally, *ab initio* VS IMSRG calculations for  $Z = 50$  have been performed and are included for comparison, showing excellent agreement with the observed behavior for  $\Delta_{2n}$  [Fig. 3(b)], while overpredicting the strength of the shell closure [Fig. 3(a)].

For the region beyond the shell closure of  $N = 82$ , the recent shell model calculations in Ref. [66] predict a decreasing  $E(2_1^+)$  energy for  $^{136-138}\text{Sn}$  isotopes and a possible new shell closure at  $^{140}\text{Sn}$  that is only obtained by including three-body interactions (whereas the effects were hindered by realistic two-body interactions). Also, the *ab initio* calculations presented indicate a subshell at  $N = 90$ ; see Fig. 3(b).

The new  $S_{2n}$  values are larger than the predictions from mass models typically used in *r*-process network calculations. The new measurements exhibit a flat trend in  $S_{2n}$  after the shell closure. If this trend continues, it would significantly shift the neutron dripline in the tin isotopic chain, affecting both the *r*-process path and overall abundance [67]. The following discussion focuses on the local impact of these new mass measurements on *r*-process calculations.

The experimental determination of nuclear masses has consistently provided new insights into *r*-process nucleosynthesis. Individual measurements affect the abundances up to a few 10s% [29,68,69]. To assess how the new mass measurements affect theoretical predictions for *r*-process nucleosynthesis, we calculated neutron capture rates for isotopes  $^{130-139}\text{Sn}$  using the TALYS statistical model code [70]. Six different sets of reaction rates are calculated. The first set employs the nuclear masses of the last AME2020 evaluation [36]. Two other sets represent the uncertainties as suggested in the AME2020, where we limit ourselves to the combination of values that maximize and minimize the  $S_{2n}$  of  $^{138}\text{Sn}$ , since we expect this to be the most relevant for nucleosynthesis. The fourth set is based on the new experimental results (see Table I), and finally two based on recent theoretical mass models, FRDM12 [40] and HFB-BSkG2 [42].

To examine the impact on the *r*-process nucleosynthesis, we utilize the reaction rates and general setup as in Ref. [61]. We use the reaction rates based on the HFB21

masses [71] as a baseline and replace the neutron ( $n, \gamma$ ) and ( $\gamma, n$ ) rates on  $^{130-139}\text{Sn}$ . We incorporate the recent experimental updates on the probabilities and rates for  $\beta$ -delayed neutron emission from [72]. We adopt the parametrization from Ref. [73] for the thermodynamic evolution. We use a specific entropy of  $s = 20k_B/\text{baryon}$  and an expansion timescale of  $\tau = 60$  ms and electron fraction  $Y_e = 0.2$ . To enhance the third  $r$ -process peak, we add 15% of material with  $Y_e = 0.15$ ,  $s = 25k_B/\text{baryon}$ , and  $\tau = 10$  ms when calculating the final abundances shown in Fig. 4. These conditions are chosen to demonstrate the impact of

the neutron capture cross sections on  $^{134-139}\text{Sn}$ , but are in agreement with recent simulations of neutron star mergers [74,75] that indicate that many  $r$ -process events might be dominated by moderately neutron-rich material with  $0.2 < Y_e < 0.3$ . Under these conditions, fission reactions do not significantly affect isotopes around the second  $r$ -process peak [61].

Figure 4 displays the resulting abundances alongside the solar  $r$ -process residuals [65]. Figure 4(a) shows the abundances when  $\beta$  decays dominate over neutron capture reactions, while Fig. 4(b) presents the final abundances. The insets in Fig. 4 highlight the mass region influenced by the new experimental data. The uncertainty based on the AME2020 extrapolation [Fig. 4(b)] mostly affects the abundances of  $A = 135$  and  $A = 137$ . The relative difference with respect to the central value of the AME2020 extrapolation is shown in Fig. 4(c). The nuclear mass uncertainties in AME2020 can lead to variations of up to  $\pm 60\%$ .

The remaining uncertainty from the new experimental values is represented as a band, which is not significantly thicker than the linewidth and is at the level of a few percent, a reduction in uncertainty by more than an order of magnitude. To understand the sources of the uncertainty when considering the masses from AME2020 (gray area), we also present the abundance pattern at the time when  $\beta$  become faster than neutron captures, shown in Fig. 4(a). Here, the AME2020 uncertainty for  $A = 138$  spans more than an order of magnitude (gray area). The very low abundance at  $A = 138$  (minimum in gray band) results from an extreme case of an almost vanishing empirical pairing gap and a minimal value of  $S_{2n}$  at  $N = 88$  allowed within the AME2020 uncertainties. This creates a larger gap in the values of  $S_{2n}$  between  $Z = 50$  and  $Z = 51$  trend lines [see Fig. 3(a)] which leads to the depletion of  $A = 138$ .

The final abundances shown in Fig. 4(b) are also influenced by  $\beta$ -delayed neutron emission. When the isotopes with  $A = 138$  and  $A = 136$  decay to stability, they encounter isotopes with a high probability of  $\beta$ -delayed neutron emission. As a result, much of the uncertainty for  $A = 136$  and  $A = 138$ , shown in Fig. 4(a), translates into uncertainty for  $A = 137$  and  $A = 135$  in Fig. 4(b). The  $\beta$ -delayed neutron emission smooths out and alters the odd-even effects along the  $r$ -process path and also somewhat reduces the sensitivity of the final abundance pattern to changes in individual nuclear masses. The final abundances calculated using theoretical mass models differ from those obtained with the new experimental results by approximately 20%.

Figure 4 shows that, under the conditions we are studying, the  $r$ -process abundance of  $^{135}\text{Ba}$  is lower than that of  $^{137}\text{Ba}$ . While the  $r$ -process contribution to these isotopes is not well constrained by [65], recent advances on  $s$ -process nucleosynthesis [76] suggest that the  $s$ -process contribution to  $^{135}\text{Ba}$  may be significantly higher than previously thought, potentially reducing the  $r$ -process residual of  $^{135}\text{Ba}$  below the value of  $^{137}\text{Ba}$ . In fact, our

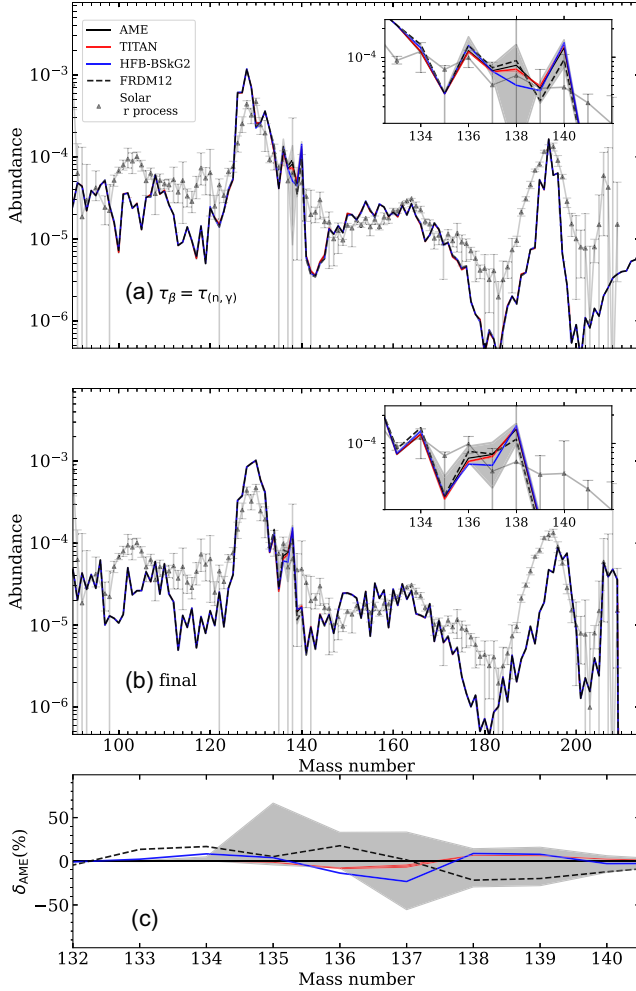


FIG. 4. (a) The calculated abundances at the time when  $\beta$  decay becomes dominant over neutron capture reactions. (b) The final abundances from  $r$ -process calculations. The insets highlight the mass region influenced by the new experimental data. The gray-shaded band represents the range of uncertainties based on AME2020, while the red curve corresponds to the experimental mass measurements. The solar  $r$ -process pattern and associated error bars [65] are scaled so that the abundances at  $A = 134$  fall within the uncertainties of the solar  $r$ -process residuals. (c) The relative difference of the final abundances compared to those based on AME2020 is shown as  $\delta_{\text{AME}} = [Y(A) - Y_{\text{AME}}(A)]/Y_{\text{AME}}(A)$ .

calculations show that a pattern with  $Y(^{137}\text{Ba}) < Y(^{135}\text{Ba})$  is clearly outside of the uncertainties determined by the nuclear masses for a broad range of conditions.

In summary, we report the successful production of neutron-rich tin isotopes at the TRIUMF facility using the newly developed  $p$ -to- $n$  target. This includes first-time mass measurements of the  $^{136}\text{Sn}$ ,  $^{137}\text{Sn}$ , and  $^{138}\text{Sn}$  isotopes, marking a significant milestone. The results highlight the potential of this target for the future mass measurements of the neighboring elements. We use these high-precision mass measurements, along with new *ab initio* calculations, to investigate the nuclear structure beyond the neutron shell closure ( $N = 82$ ). Our experimental and theoretical findings reveal a notable decrease in the empirical pairing gap well beyond  $N = 82$ . This behavior is poorly reproduced by the nuclear mass models such as FRDM, HFB21, and HFB-BSkG2 within their expected uncertainties.

The new mass measurements have expanded our knowledge of the  $S_{2n}$  region, revealing a flatter trend compared to heavier elements. If the observed flat trend continues, it would shift both the  $r$ -process path and the neutron dripline in tin toward heavier species [67].

The new measurements significantly improve the uncertainty of the mass input in a key region for the  $A = 130$  abundance peak. With the new experimental results, our  $r$ -process calculations also predict a lower abundance of  $A = 135$  compared to  $A = 137$  in the chosen scenario, which supports the predicted higher contribution to the  $s$  process for  $^{135}\text{Ba}$ .

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