Atomic masses of nuclei with neutron numbers $N < 126$ and proton numbers $Z > 82$

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In this paper we study atomic masses of the major shells to the northwest of the ^{208}Pb nucleus, namely, those with neutron numbers $N < 126$ and proton numbers $Z > 82$, in terms of neutron-proton interactions. With statistical corrections, we are able to construct mass formulas for nuclei in this region with root-mean-squared deviation close to 30 keV, which is more accurate than all previous efforts. Predicted results of some unaccessible masses are tabulated in the Supplemental Material of this article.

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I. INTRODUCTION

Nuclear mass is one of the fundamental properties of an atomic nucleus. There have been continuous efforts towards understanding this quantity. Atomic mass evaluation of experimental measurements has been reviewed every four or five years, and the latest three versions are the AME2012 [1], AME2016 [2], and AME2021 [3] databases. Theoretically, a few mass models were established, e.g., the Duflo-Zuker model [4], the Skyrme-Hartree-Fock Bogoliubov theory (SHFB) [5], the Weizsäcker-Skyrme model [6], and the finite-range droplet model (FRDM) [7]. Besides these theoretical models, local mass relations have been proved to be useful, and efforts along this line include the Audi-Wapstra extrapolation method [8], the Garvey-Kelson (G-K) mass relations [9–13], mass formulas related to the neutron-proton interaction relation [14,15], and mass formulas between mirror nuclei [16–18].

The purpose of this paper is to report mass formulas for nuclei in the major shells with neutron numbers $N < 126$ and proton numbers $Z > 82$, namely, where both valence protons and valence neutrons are in the 82–126 major shells, or northwest of the doubly-closed-shell ^{208}Pb nucleus in the nuclear chart. For convenience, we treat neutrons as holes with respect to the $N = 126$ major shell. With statistical correlations taken into account, we are able to construct simple formulas of the neutron-proton interaction between the last i neutrons and the last j protons, denoted by $\delta V_{in\text{-}jp}$, within the root-mean-squared deviation close to 30 keV, and based on which we demonstrate the predictive power of these formulas, which have a remarkable accuracy.

This paper is organized as follows: In Sec. II we study a linear dependence of $\delta V_{1n\text{-}1p}$ on A, and study the statistical linear correlation of deviations of calculated $\delta V_{1n\text{-}1p}$ from experimentally defined $\delta V_{1n\text{-}1p}$, where mass formulas are obtained with an accuracy typically about 30 keV. In Sec. III we apply our formulas and make predictions of nuclear masses which are not accessible in the AME2021 database. In Sec. IV there is a brief summary.

II. NEUTRON-PROTON INTERACTIONS AND MASS FORMULAS

The important role of the neutron-proton interaction in the evolution of collectivity, deformation, and phase transitions was stressed a long time ago by Shalit and Goldhaber [19] and Talmi [20]. A surrogate of neutron-proton interaction strength is the empirically obtained interaction for even-even nuclei. This neutron-proton interaction was first constructed and studied by Zhang, Brenner, Casten, and collaborators [21,22] and later refined by Fu and collaborators [23]. An integrated neutron-proton interaction $V_{\rm NP}$ was studied in Refs. [21,24], which exhibits a linearity with N_nN_p, the product of valence neutron and proton numbers. Casten and Zamfir proposed and studied extensively the evolution of nuclear structure in terms of the simple N_nN_p scheme [25].

The systematics of the neutron-proton interaction was applied to mass formulas in successive papers by Fu, Jiang, and collaborators [14,15,26]. They defined the empirical neutron-proton interaction between the last i neutrons and j protons, denoted by $\delta V_{in\text{-}jp}$ as

$$\delta V_{in\text{-}jp}(N,Z) = M(N,Z) + M(N-i,Z-j) - M(N-i,Z) - M(N,Z-j), \qquad (1)$$

where $M(N,Z)$ is the atomic mass of a nucleus with N neutrons and Z protons. Apparently, once $\delta V_{in\text{-}jp}$ is available, one immediately obtains relations of four neighboring nuclei with neutron and proton numbers (N,Z), $(N-i,Z-j)$, $(N-i,Z)$, and $(N,Z-j)$. Therefore, it is highly desirable to evaluate the value of $\delta V_{in\text{-}jp}$ as accurate as possible, in advance. Usually, $\delta V_{in\text{-}jp}$ is given by empirical formulas which

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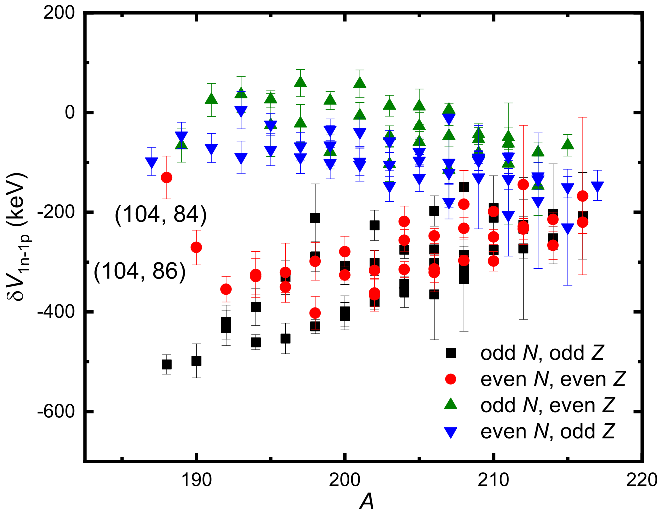

FIG. 1. $\delta V_{1n\text{-}1p}$, extracted based on the AME2021 database [3], vs mass number A, for nuclei with neutron numbers $N < 126$ and proton numbers $Z > 82$.

yield root-mean-squared deviations (RMSD) about 130–170 keV for mass number $A \geqslant 100$, in comparison with those extracted from experimental data.

In Ref. [27], it was reported that the neutron-proton interaction is very small when both the number of valence protons and the number of valence neutron holes for nuclei in the major shells to the northwest of the ^{208}Pb nucleus are small. This is actually understandable, because in this case valence protons are in the high-j $h_{9/2}$ orbit and valence neutron holes are in the low-j, successively, $p_{1/2}$, $p_{3/2}$, and $f_{5/2}$ orbits, and thus the neutron-proton interaction strength is very small due to the small overlaps between those valence protons and neutron holes. As the numbers of valence protons and neutron holes increase, more and more configurations involving of the f orbits for neutron holes set in, the overlap between valence protons and neutron holes increases, and so does the neutron-proton interaction. From another perspective, proton separation energy and neutron separation energy exhibit a linear combination of N_p and N_n [28], and this linearity is excellent in particular in this region [29]. Because $\delta V_{1n\text{-}1p}$ could be written as the difference between two separation energies, the value of $\delta V_{1n\text{-}1p}$ is expected to evolve very smoothly and compactly throughout the valence shells to the northwest of ^{208}Pb.

In Fig. 1, we plot $\delta V_{1n\text{-}1p}$, extracted by using the AME2021 database [3], versus mass number A for experimentally accessible nuclei in this region, as an exemplification of $\delta V_{in\text{-}jp}$. One sees that the magnitude of $\delta V_{1n\text{-}1p}$ of nuclei with even value of mass number A is very small indeed, although with fluctuations, for those nuclei close to ^{208}Pb, and becomes larger for those with smaller A (note that the $\delta V_{1n\text{-}1p}$ of nuclei with even values of A is negative). As for $\delta V_{1n\text{-}1p}$ of nuclei with odd A, in Ref. [14] the value of $\delta V_{1n\text{-}1p}$ was assumed to be a constant value (−74 keV) for all nuclei with mass number $A \geqslant 100$, and the odd-even difference of $\delta V_{1n\text{-}1p}$ was interpreted in terms of a subtle effect of pairing interaction. One sees in Fig. 1 that the values of $\delta V_{1n\text{-}1p}$ for the odd-A case exhibit a compact trajectory which decreases very slightly with A. This behavior brings a correction to the simple relation $\delta V_{1n\text{-}1p} \simeq -74$ keV (the RMSD of which is −60 keV), and thus is expected to yield a more accurate formula of $\delta V_{1n\text{-}1p}$ (to be explained later in this paper) than the previous relation $\delta V_{1n\text{-}1p} \simeq -74$ keV.

TABLE I. Parameters C and a in Eq. (2) and corresponding RMSDs (denoted by σ) of $\delta V_{in\text{-}jp}$ in keV, the so-called Pearson correlation coefficient r between $\Delta V_{in\text{-}jp}(N, Z)$ and $\Delta V_{in\text{-}jp}(N-2, Z-2)$, the linear coefficient λ in Eq. (4), and the RMSD value σ' with the improvement of statistical linear correlation described by Eq. (4), for nuclei with neutron numbers $N < 126$ and proton numbers $Z > 82$.

	C	a	σ	λ	r	σ'
$\delta V_{1n\text{-}1p}$ (odd A)	−142(16)	−4.25(88)	52	0.73 (6)	0.81	26
$\delta V_{1n\text{-}1p}$ (even A)	−179(19)	7.14(107)	65			34
$\delta V_{1n\text{-}2p}$	−328(14)	2.32(83)	62	0.87 (6)	0.87	29
$\delta V_{2n\text{-}1p}$	−364(16)	−0.44(101)	70	0.77 (6)	0.85	35
$\delta V_{2n\text{-}2p}$	−642(17)	6.14(105)	75	0.88 (7)	0.84	37

The values of $\delta V_{in\text{-}jp}$ with $(i, j) = (1, 2)$, $(2, 1)$, and $(2, 2)$ exhibit a similar pattern as $\delta V_{1n\text{-}1p}$, but without odd-even discrimination. A simple formula of $\delta V_{in\text{-}jp}$ with this property is given as

$$\delta V^{\rm cal}_{in\text{-}jp}(N, Z) = C + a(A - 220), \tag{2}$$

where C and a are free parameters ($\delta V_{1n\text{-}1p}$ is discriminated between odd A and even A cases). Here the constant 220 is adopted artificially, as the largest mass in this region is close to this number; without this constant, all values of C would be −1000 keV or even lower. The optimized C and a, together with corresponding RMSD, are presented in the first three columns of Table I, according to which the root-mean-squared deviation, denoted by σ, between $\delta V_{in\text{-}jp}$ calculated by using Eq. (2) and those extracted from the AME2021 database changes fluctuates between 50 to 75 keV, with σ of $\delta V_{1n\text{-}1p}$ being the smallest.

In order to further reduce the RMSD values of our calculated $\delta V_{1n\text{-}1p}$ by using Eq. (2), we introduce a quantity

$$\Delta V_{in\text{-}jp}(N, Z) = \delta V_{in\text{-}jp}(N, Z) - \delta V^{\rm cal}_{in\text{-}jp}(N, Z). \tag{3}$$

The first term on the right-hand side of the above formula, $\delta V_{in\text{-}jp}$, is calculated by using Eq. (1) in terms of experimental data, and the second term, $\delta V^{\rm cal}_{in\text{-}jp}$, is calculated by using Eq. (2) with parameters in Table I. In Fig. 2, we plot the correlation between $\Delta V_{1n\text{-}1p}(N, Z)$ and $\Delta V_{1n\text{-}1p}(N-2, Z-2)$. Apparently, these two quantities exhibit statistical correlations; without details we note that other $\Delta V_{in\text{-}jp}$ exhibit similar patterns. We thus assume

$$\Delta V_{in\text{-}jp}(N, Z) = \lambda \Delta V_{in\text{-}jp}(N-2, Z-2), \tag{4}$$

where λ is the linear-correlation coefficient, listed in the fourth column of Table I, together with the so-called Pearson correlation coefficient (denoted by r, listed in the fifth column of Table I). If the value of r is larger than 0.8, usually, one calls the correlation strong. Therefore according to Table I, all these linear correlations of $\Delta V_{in\text{-}jp}(N, Z)$ versus $\Delta V_{in\text{-}jp}(N-2, Z-2)$ are strong.

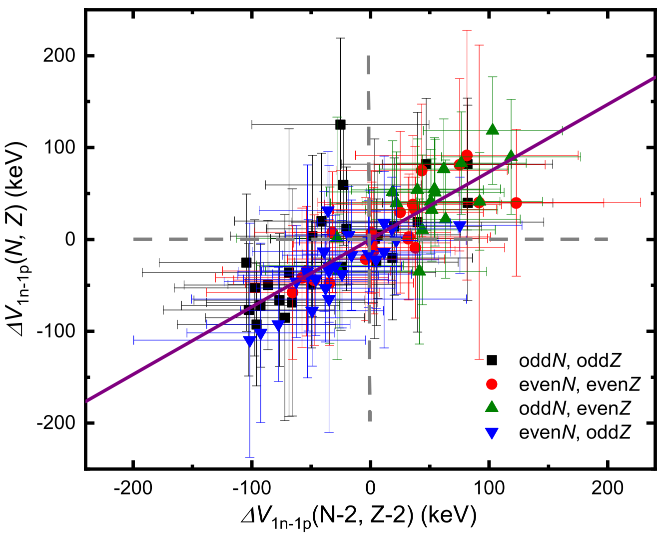

FIG. 2. $\Delta V_{1n\text{-}1p}(N,Z)$ versus $\Delta V_{1n\text{-}1p}(N-2,Z-2)$, defined in Eq. (3). The solid straight line in purple is plotted by the optimal value of λ.

With this statistical correlation, the refined formula for our theoretically predicted $\delta V_{in\text{-}jp}$, denoted by $\delta V^{\rm th}_{in\text{-}jp}$, is

$$\delta V^{\rm th}_{in\text{-}jp}(N,Z) = \delta V^{\rm cal}_{in\text{-}jp}(N,Z) + \lambda \Delta V_{in\text{-}jp}(N-2,Z-2). \tag{5}$$

On the right-hand side of the above formula, the first term $\delta V^{\rm cal}_{in\text{-}jp}$ is calculated by using Eq. (2), the parameter λ in the second term is taken from Table I, and the second term $\Delta V_{in\text{-}jp}$ is calculated by using Eq. (3). We denote the RMSD of the above formula by σ' and tabulate them in the last column of Table I. One sees substantial improvements by considering this statistical correlation; here the RMSD values are reduced by about 50%.

Besides the above formulas of $\delta V_{in\text{-}jp}$, in Ref. [30] a number of local mass formulas were unified in the form of $S^{(i,j)}_{N',Z'}(N,Z) \simeq 0$ (including the nice relation of α-decay energies of neighboring nuclei reported in Ref. [31]), where

$$S^{(i,j)}_{N',Z'}(N,Z) = \delta V_{in\text{-}jp}(N,Z) - \delta V_{in\text{-}jp}(N-N',Z-Z'). \tag{6}$$

We note without details that similar correlation exists for these mass relations; unfortunately, their RMSD values are systematically larger than those in Table I.

III. APPLICATIONS OF OUR MASS FORMULAS

In this section, we apply our approach of $\delta V_{in\text{-}jp}$ in Eq. (5) and predict atomic masses which are not accessible in the AME2021 database in these major shells.

The procedure adopted in this paper is explained as follows. We make use of our theoretically predicted $\delta V^{\rm th}_{in\text{-}jp}$ in the extrapolation of atomic masses. Replacing $\delta V_{in\text{-}jp}$ in Eq. (1) by using $\delta V^{\rm th}_{in\text{-}jp}$, we have

$$\delta V^{\rm th}_{in\text{-}jp}(N,Z) = M(N,Z) + M(N-i,Z-j) - M(N-i,Z) - M(N,Z-j).$$

From the above formula, we obtain our predicted masses, $M^{\rm pred}(N,Z)$, via extrapolations:

$$\begin{aligned} M^{\rm pred}(N,Z) &= \delta V^{\rm th}_{in\text{-}jp}(N,Z) - M(N-i,Z-j) \\ &\quad + M(N-i,Z) + M(N,Z-j), \\ M^{\rm pred}(N,Z) &= \delta V^{\rm th}_{in\text{-}jp}(N+i,Z+j) - M(N+i,Z+j) \\ &\quad + M(N+i,Z) + M(N,Z+j), \\ M^{\rm pred}(N,Z) &= -\delta V^{\rm th}_{in\text{-}jp}(N+i,Z) - M(N+i,Z-j) \\ &\quad + M(N+i,Z) + M(N,Z-j), \\ M^{\rm pred}(N,Z) &= -\delta V^{\rm th}_{in\text{-}jp}(N,Z+j) - M(N-i,Z+j) \\ &\quad + M(N,Z+j) + M(N-i,Z). \end{aligned} \tag{7}$$

For one-proton separation energy S_p, we have

$$\delta V^{\rm th}_{in\text{-}jp}(N,Z) = S_p(N-i,Z) - S_p(N,Z).$$

From this formula, we obtain

$$\begin{aligned} S^{\rm pred}_p(N,Z) &= S_p(N-1,Z) - \delta V^{\rm th}_{1n\text{-}1p}(N,Z), \\ S^{\rm pred}_p(N,Z) &= S_p(N+1,Z) + \delta V^{\rm th}_{1n\text{-}1p}(N+1,Z), \\ S^{\rm pred}_p(N,Z) &= S_p(N-2,Z) - \delta V^{\rm th}_{2n\text{-}1p}(N,Z), \\ S^{\rm pred}_p(N,Z) &= S_p(N+2,Z) + \delta V^{\rm th}_{2n\text{-}1p}(N+2,Z). \end{aligned} \tag{8}$$

Similarly, for two-proton separation energy S_{2p},

$$\begin{aligned} S^{\rm pred}_{2p}(N,Z) &= S_{2p}(N-1,Z) - \delta V^{\rm th}_{1n\text{-}2p}(N,Z), \\ S^{\rm pred}_{2p}(N,Z) &= S_{2p}(N+1,Z) + \delta V^{\rm th}_{1n\text{-}2p}(N+1,Z), \\ S^{\rm pred}_{2p}(N,Z) &= S_{2p}(N-2,Z) - \delta V^{\rm th}_{2n\text{-}2p}(N,Z), \\ S^{\rm pred}_{2p}(N,Z) &= S_{2p}(N+2,Z) + \delta V^{\rm th}_{2n\text{-}2p}(N+2,Z), \end{aligned} \tag{9}$$

except for $Z=83$ for which the values of $\delta V^{\rm th}_{1n\text{-}2p}$ and $\delta V^{\rm th}_{2n\text{-}2p}$ involve nuclei beyond the major shells to the northwest of ^{208}Pb; for this case the value of S_{2p} is evaluated by

$$S_{2p}(N,Z) = -M^{\rm pred}(N,Z) + M^{\rm exp}(N,Z-2) + 2M(^1{\rm H}).$$

Eqs. (7)–(9) are applied recursively in our extrapolation process.

The uncertainty of our predicted results is taken to be the squared root of a sum over uncertainties squared of all terms on the right-hand side in a given formula. For instance, the uncertainty of $M^{\rm pred}(N,Z)$ in the first formula of Eq. (7) is given by

$$\begin{aligned} [\sigma_{\rm pred}(N,Z)]^2 &= [\sigma_{\rm th}(N,Z)]^2 + [\sigma_{\rm exp}(N-i,Z-j)]^2 \\ &\quad + [\sigma_{\rm exp}(N-i,Z)]^2 \\ &\quad + [\sigma_{\rm exp}(N,Z-j)]^2, \end{aligned} \tag{10}$$

where $\sigma_{\rm th}$ is given by σ' in Table I, and $\sigma_{\rm exp}$ is adopted from the AME2021 database.

If there are two or more channels to predict one quantity, the predicted result is simply taken to the average weighted by their accuracies, as in the procedure in the literature, e.g., Ref. [14]: Suppose that a given mass M has two predicted results, $M^{\rm pred1} = M^{(1)} \pm \sigma_1$ and $M^{\rm pred2} = M^{(2)} \pm \sigma_2$;

TABLE II. Predicted mass excesses (not accessible in the AME2012 database but available in the AME2021 database for nuclei with neutron numbers $N < 126$ and proton numbers $Z > 82$) and their uncertainties (in keV) predicted in this work, together with predictions by using the approaches of Refs. [15,32] with readjusted parameters, and predictions in Ref. [6]. The corresponding RMSD values are tabulated in the last row (values inside the bracket are RMSD values by excluding ^{219}Np). One sees the predicted results in this work yield the smallest RMSD among all, with respect to the AME2021 database.

Element	AME2021	Jiang *et al.*	Ma *et al.*	WS4+RBF	This work
^{198}At	-6708 ± 4	-6635 ± 60	-6714 ± 69	-6971	-6611 ± 30
^{197}Fr	10253 ± 56	10440 ± 117	10334 ± 60	10282	10409 ± 47
^{198}Fr	9577 ± 31	9597 ± 90	9513 ± 84	9183	9598 ± 47
^{202}Fr	3101 ± 6	3123 ± 69	3056 ± 41	2876	3094 ± 23
^{201}Ra	11936 ± 20	12033 ± 100	11970 ± 73	11819	11883 ± 56
^{205}Ac	14106 ± 59	14049 ± 119	14031 ± 62	13972	14073 ± 47
^{206}Ac	13484 ± 65	13391 ± 94	13375 ± 82	13249	13465 ± 28
^{211}Pa	22052 ± 69	22020 ± 94	22002 ± 56	21877	22077 ± 52
^{215}U	24889 ± 104	25183 ± 128	24817 ± 62	25212	24844 ± 51
^{216}U	23066 ± 28	23282 ± 130	23072 ± 56	23118	23027 ± 44
^{219}Np	29436 ± 91	29606 ± 249	29114 ± 153	29316	-
RMSD(keV)		143 (140)	114 (62)	207 (214)	58

the final result in this case is given by $M^{\text{pred}} = M^{\text{th}} \pm \sigma^{\text{th}}$, with

$$M^{\text{th}} = \frac{\frac{M^{(1)}}{(\sigma_1)^2} + \frac{M^{(2)}}{(\sigma_2)^2}}{\frac{1}{(\sigma_1)^2} + \frac{1}{(\sigma_2)^2}}, \qquad \frac{1}{(\sigma^{\text{th}})^2} = \frac{1}{(\sigma_1)^2} + \frac{1}{(\sigma_2)^2}.$$

Let us now demonstrate the predictive power of this method, by a comparison of our predicted results not accessible in the AME2012 database [1] with those accessible in the AME2021 database [3]. To be precise, we note that we have adopted the extrapolated result of the ^{194}Bi nucleus in the AME2012 database, in order to predict the masses of ^{198}At, ^{202}Fr and ^{206}Ac; the reason is that the key formula, Eq. (5),

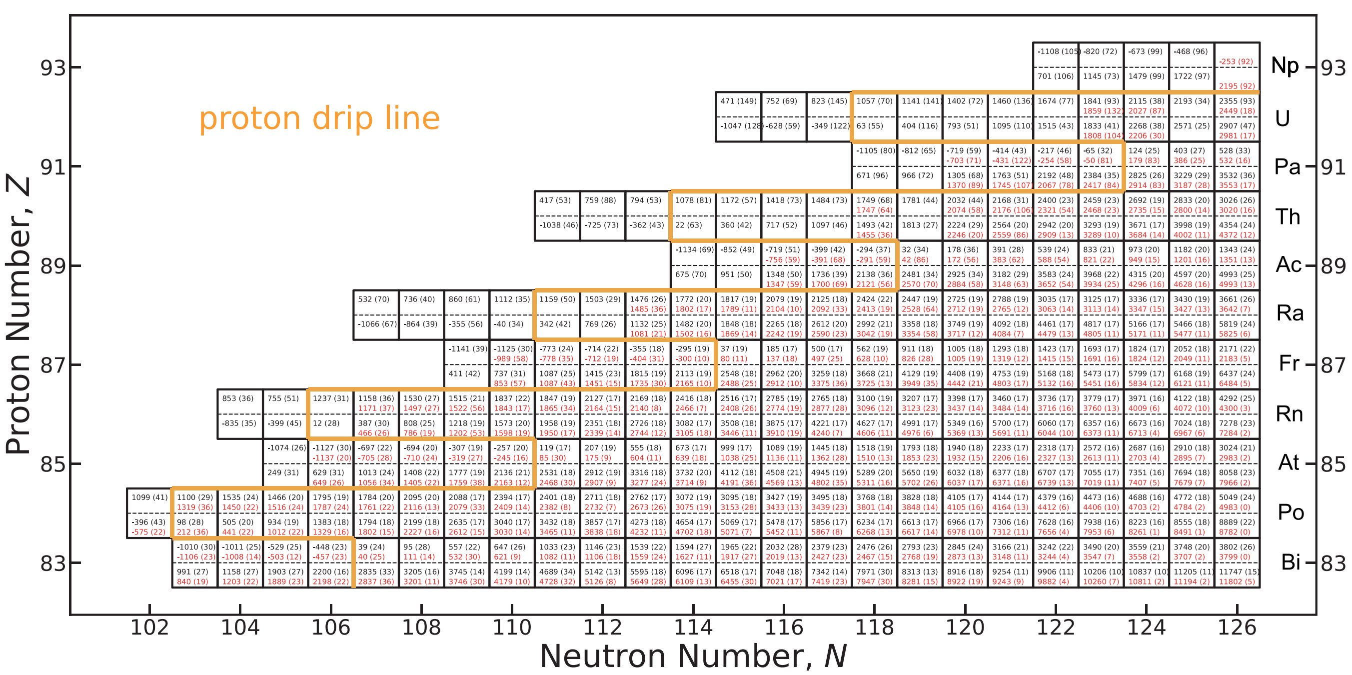

FIG. 3. Predicted one- and two-proton separation energies (S_p and S_{2p}, in keV) for nuclei with proton number Z from 83 to 93. In each block, the first row corresponds to S_p predicted in this work, the second row corresponds to S_p in Ref. [3], the third row corresponds to S_{2p} predicted in this work, and the fourth row corresponds to S_{2p} in Ref. [3]. The predicted results in this work and experimental data in Ref. [3] are in black and red, respectively. The values in the brackets correspond to uncertainties. The orange solid line plots the proton drip line. We note that the extrapolated results of the ^{186}Po nucleus could be problematic, because theoretical $\delta V_{1n\text{-}1p}$ for one of its neighboring nuclei, ^{188}Po, deviates sizably from experimental data.

of our extrapolation process involves of a correlation between two nuclei, one with neutron and proton numbers (N, Z) and the other with $(N + 2, Z + 2)$; it takes one step from ^{194}Bi to ^{198}At, one step from ^{198}At to ^{202}Fr, and one step from ^{202}Fr to ^{206}Ac.

The results of our numerical experiment are partially tabulated in Table II, in which we also present the predicted results of a few other approaches in the literature, with parameters of Refs. [15,32] readjusted by using the AME2012 database. One sees that the predicted results in this paper are in best agreement with those in the latest AME2021 database. As additional evidence of the strong predictive power in this region, we predict the atomic mass of ^{214}U. The latest experimental result of alpha-particle emission energy E_α of ^{214}U in Ref. [33], 8533(18) keV, yields an alpha-decay energy of 8696 (18) keV. Our predicted result is 8654(47) keV (see the Supplemental Material of this paper [34]), which is again remarkably consistent with the result of experimental measurement.

In Fig. 3 we present our predicted results of one- and two-proton separation energies larger than −1.2 MeV, based on the AME2021 database for proton numbers between 83 and 93, for the sake of convenience. Our predicted atomic mass excesses, one- and two-proton separation energies, and α-decay energies, for systems with proton number 83–93 and neutron number 104–125, are tabulated in the Supplemental Material [34] (We note that the predicted results of the ^{186}Po nucleus might be problematic and could be put in doubt, because the theoretical value of $\delta V_{1n\text{-}1p}$ involving a neighboring nucleus, ^{188}Po, deviates sizably from that extracted from experimental data).

IV. SUMMARY

In this paper, we investigate the residual proton-neutron interaction $\delta V_{1n\text{-}1p}$ for neutron numbers $N < 126$ and proton numbers $Z > 82$. Empirical formulas of neutron-proton interactions $\delta V_{in\text{-}jp}$ are reported to be, despite fluctuations, linear with mass number A in this region. With corrections of statistical correlation, the RMSD values of these formulas are reduced to 26–37 keV, which is more accurate than all previous formulas for nuclear masses. According to our numerical experiments, an application of the same procedure to other regions, e.g., the region northeast of ^{208}Pb, yields considerably larger uncertainties, unfortunately.

We demonstrate the predictive power of our formulas by using numerical experiments of extrapolation from the AME2012 database to the AME2021 database. We find that the present approach, with simplicity, yields the best consistency between predicted results and experimental data not accessible in the AME2012 database but accessible in the AME2021 database.

Finally we present our predicted masses, one- and two-proton separation energies, and α-decay energies in the Supplemental Material [34].

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[34] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevC.105.064304 for predicted mass excesses and related quantities for systems with neutron numbers $N < 126$ and proton numbers $Z > 82$.