Prediction on the heaviest proton emitters

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Based on the $Q_{p/\alpha}$ values deduced from the linear extrapolations along isotopic chains and on the universal decay law, the proton- and α-decay partial half-lives are calculated for odd-Z, even-N neutron-deficient Bi-Pa isotopes. Eight proton-emission states in five new isotopes are suggested, including the $1/2^+$ and $9/2^-$ states in ^{183}Bi, the $1/2^+$ and $7/2^-$ states in ^{187,189}At, the $1/2^+$ state in ^{193}Fr and the $9/2^-$ state in ^{199}Ac. The calculated half-lives for the $1/2^+$ states in ^{183}Bi and ^{187}At are around 100 ns, too short to be studied using the recoil separator setups, which strongly encourages the development of new experimental techniques and devices to search for new sub-microsecond proton-emitting nuclei.

I. Introduction

The proton radioactivity, whereby the proton is emitted from the nucleus, is a well-known decay mode. Two types of the proton radioactiviy are usually considered: direct proton emission from the ground state (gs) or isomer [1], and β-delayed proton emission [2]. This work deals with the first type, which occurs beyond the proton drip line and establishes the limits of existence for the majority of neutron-deficient isotopic chains. It is a key, and often the only source of information on nuclear structure and the mass surface in the most neutron-deficient region in the chart of nuclides [1, 3–6]. Therefore, predictions on the existence and properties of proton emission hold significant scientific implications, e.g., validation of theoretical models [7, 8] and inspiring experimental research [9].

Although the theoretical concept of proton emission was proposed in 1960s [10], the first evidence came only in early 1970s, when a weak proton emission branch was observed from the $19/2^-$ isomeric state ^{53m}Co [11–13]. In the early 1980s, the first ground-state (gs) proton emitter ^{151}Lu was reported [14]. Two most recent examples, proton-emitting nuclei ^{149}Lu and ^{116}La, were reported in 2022 [15, 16]. So far, 33 proton emitters have been reported for odd-Z elements between $53 \leq$ Z ≤ 83 except promethium ($Z = 61$), see reviews in Refs. [1, 4–6, 8]. Among these proton-emitting nuclei, ^{149}Lu is the shortest-lived proton emitter with $Q_p =$ 1920(20) keV and $T^p_{1/2} = 470^{+170}_{-100}$ ns [15]. The ^{185}Bi (Z=83, N=102) with the gs half-life of $2.8^{+2.3}_{-1.0}$ μs is the heaviest proton emitter and the only known one above the $Z = 82$ shell closure, which was discovered nearly 30 years ago [17, 18], with the recent investigation [19] solving a number of puzzles in its previously reported properties.

The present study focuses on predicting the new proton emitters above the $Z = 82$ shell closure and calculating the corresponding partial proton-decay half-lives. In the neutron-deficient region above $Z = 82$, the major competitive decay mode to proton emission is α decay, as shown in Fig. 1. Therefore, in order to predict the proton radioactivity of unknown nuclides, it is necessary to theoretically calculate the partial half-lives for both proton emission and α decay. Historically, many macroscopic and semi-empirical models have been developed to investigate the α-decay and/or proton-emission probabilities, such as the pre-formation cluster model [20], the unified fission model [21], the effective liquid drop model [22], the generalized liquid drop model [23], the Coulomb and proximity potential model [24], semi-empirical Geiger-Nuttal law [25] for α decay and Geiger-Nuttal-like law [26] for proton emission. The microscopic approaches for charged-particle emission include such as the R-matrix theory as formulated by Teichman and Wigner [27], the semi-empirical universal decay law (UDL) [28, 29] within the R-matrix framework, two-potential approach with Skyrme-Hartree-Fock [30], and shell model treatment involving BCS approach [31, 32]. It is worth noting that among these models, the UDL approach can simultaneously describe proton- and α-decay half-lives, by accounting for the released energy $Q_{p/\alpha}$ in decay and the orbital angular momentum $l_{p/\alpha}$ carried by the emitted particle. Therefore, in this work we used the UDL framework to calculate the half-lives for the nuclei of interest and to predict the new proton-emitting candidates.

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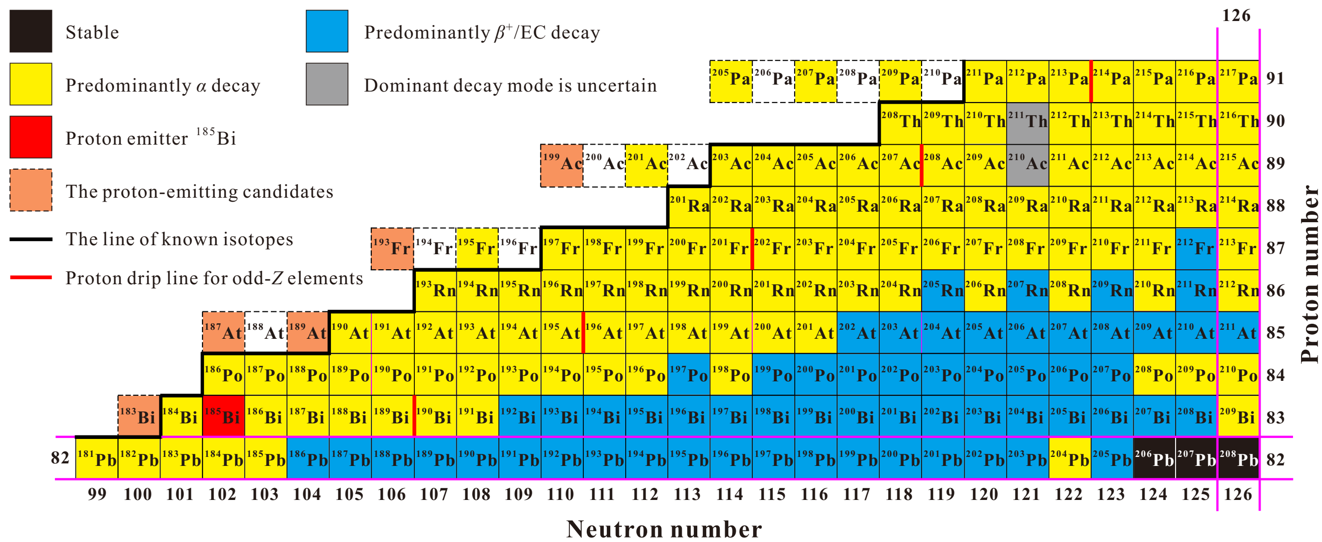

FIG. 1. The partial chart of nuclides for the Pb-Pa region. The candidates for proton emitters are marked by the brown squares, as explained in the top right-hand side corner of the plot.

Compared to α decay, the proton-decay half-life is highly sensitive to the orbital angular momentum l_p and and the released energy Q_p. The former is because, for a given mother nucleus and an orbital angular momentum carried by proton/α, the centrifugal barrier in proton emission is approximately four times higher that of α decay. The latter is empirical: for known proton emitters with $N \geq 82$ and with the same spin-parity, half-lives drop by roughly one order of magnitude for every 100-keV increase in Q_p. Meanwhile, for typical α-decaying nuclei above lead and with s-wave emission ($l_\alpha = 0$), half-lives decreases by roughly one order of magnitude for about every 300-keV increase in Q_α. As the global mass formulae [33–37] typically have a root-mean-square deviation (RMSD) of several hundred keV, using them to deduce Q_p and subsequently calculate $T^p_{1/2}$ is of limited significance. In the present work, a local linear extrapolation method is used to determine the $Q_{p/\alpha}$ for nuclei that are unknown but of interest.

Through the linear extrapolated $Q_{p/\alpha}$ along isotopic chains and the calculated $T^{p/\alpha}_{1/2}$ by the UDL, we can suggest eight proton-emission candidate states in five new isotopes above the $Z = 82$ shell closure, see Fig. 1. These candidates are $1/2^+$ and $9/2^-$ states in ^{183}Bi, $1/2^+$ and $7/2^-$ states in ^{187,189}At, $1/2^+$ state in ^{193}Fr and $9/2^-$ state in ^{199}Ac, and the selection criterion for them are described in Sec. IV.

II. Estimation of $Q_{p/\alpha}$ values

In order to predict new proton-emitting nuclei above the Z=82 shell closure, the proton-separation energies for known neutron-deficient isotopes of odd-Z elements from Bi (bismuth) to Pa (protactinium) are investigated [38]. In this region, proton emission cannot compete with α decay until at least several mass units beyond the drip line. For example, in the case of ^{185}Bi, this occurs already 5 mass units beyond the proton drip line, as shown in Fig. 1. Considering the complex proton-neutron multiplets found in odd-odd nuclei, only odd-Z, even-N nuclei are investigated in the present work.

We begin our analysis by considering the systematics of Q_p and Q_α for a given state in an isotopic chain where three or more consecutive data were known. As an example, the upper panel of Fig. 2 shows the Q_p values for the $1/2^+$ states in odd-A $^{185-193}$Bi isotopes and the corresponding linear fitting, and the corresponding residuals are displayed in the lower panel with an RMSD of 12.9 keV. This result confirms a linear trend of Q_p in odd-A $^{185-193}$Bi isotopes. The similar linear trends can be seen in both Q_p and Q_α for a given state of At, Fr, Ac and Pa isotopic chains, as shown in Supplemnental Material and indicating good linearity for both Q_p and Q_α in this region. It is worth noting that although the macroscopic-microscopic model predicts a gs spherical-oblate-prolate shape transition between N = 100-120 in this region [40], it appears that this shape transition has no significant affect on the linearity of Q_p and Q_α.

Based on this linear trend of Q_p and Q_α, we assume that linearity still holds for at least a limited region of lighter isotopes in each chain. Thus, a linear extrapolation approach is used, and $Q_{p/\alpha}$ values were estimated for at most three unknown isotopes in an isotopic chain. Table I shows the known data [38, 39] and the linear extrapolation values of Q_p and Q_α. Based on the known data and the linear extrapolation assumption, the errors of Q_p and Q_α values for unknown isotopes can

TABLE I. The known data [38, 39] and linear extrapolated (in bold) Q_p and Q_α values for odd-Z, even-N neutron-deficient Bi-Pa isotopes. Tentative spin-parity assignments proposed in the literatures are given in parentheses. The spin-parities for the predicted proton-emitting candiates are based on systematics, see discussion in Sec. III A

Nuclides J^π	Q_p/keV	Q_α/keV	Nuclides J^π	Q_p/keV	Q_α/keV	Nuclides J^π	Q_p/keV	Q_α/keV
^{193}Bi (1/2$^+$)	-317(11)	6612(13)	^{197}At (1/2$^+$)	-123(14)	6846(4)	^{205}Fr (1/2$^+$)	-20(13)	7205(6)
^{191}Bi (1/2$^+$)	128(15)	7020(12)	^{195}At (1/2$^+$)	245(16)	7098(5)	^{203}Fr 1/2$^+$	223(20)	7392(5)
^{189}Bi (1/2$^+$)	641(25)	7452(30)	^{193}At (1/2$^+$)	710(24)	7388(5)	^{201}Fr 1/2$^+$	429(15)	7608(9)
^{187}Bi (1/2$^+$)	1121(25)	7891(24)	^{191}At (1/2$^+$)	1138(21)	7708(11)	^{199}Fr (1/2$^+$)	713(50)	7821(11)
^{185}Bi 1/2$^+$	1607(20)	8218(18)	**^{189}At 1/2$^+$**	**1555(29)**	**7979(14)**	**^{197}Fr 1/2$^+$**	**987(66)**	**8023(13)**
^{183}Bi 1/2$^+$	**2088(26)**	**8664(31)**	**^{187}At 1/2$^+$**	**1979(39)**	**8267(27)**	**^{195}Fr 1/2$^+$**	**1243(89)**	**8229(18)**
^{195}Bi (9/2$^-$)	-1107(18)	5535(8)	^{195}At (7/2$^-$)	275(19)	7223(4)	**^{193}Fr 1/2$^+$**	**1498(112)**	**8435(23)**
^{193}Bi (9/2$^-$)	-622(9)	6026(5)	^{193}At (7/2$^-$)	715(26)	7480(5)	^{209}Fr 9/2$^-$	-1402(18)	6777(4)
^{191}Bi (9/2$^-$)	-112(15)	6441(3)	^{191}At (7/2$^-$)	1193(37)	7817(15)	^{207}Fr 9/2$^-$	-1018(23)	6893(20)
^{189}Bi (9/2$^-$)	457(23)	6816(3)	**^{189}At 7/2$^-$**	**1646(43)**	**8101(30)**	^{205}Fr (9/2$^-$)	-629(11)	7055(2)
^{187}Bi (9/2$^-$)	1010(15)	7154(5)	**^{187}At 7/2$^-$**	**2105(62)**	**8398(68)**	^{203}Fr (9/2$^-$)	-138(19)	7274(5)
^{185}Bi 9/2$^-$	**1548(28)**	**7549(6)**	^{215}Pa (9/2$^-$)	-180(80)	8240(7)	^{201}Fr (9/2$^-$)	300(11)	7510(7)
^{183}Bi 9/2$^-$	**2095(38)**	**7925(8)**	^{213}Pa 9/2$^-$	250(60)	8394(15)	**^{199}Fr 9/2$^-$**	**773(29)**	**7735(12)**
^{213}Ac 9/2$^-$	-949(16)	7498(4)	^{211}Pa (9/2$^-$)	700(70)	8480(40)	**^{197}Fr 9/2$^-$**	**1238(42)**	**7962(16)**
^{211}Ac 9/2$^-$	-550(60)	7620(50)	**^{209}Pa 9/2$^-$**	**1084(86)**	**8611(61)**	**^{195}Fr 9/2$^-$**	**1702(55)**	**8190(21)**
^{209}Ac (9/2$^-$)	-160(50)	7730(50)	**^{207}Pa 9/2$^-$**	**1493(112)**	**8731(88)**			
^{207}Ac (9/2$^-$)	290(60)	7845(25)	**^{205}Pa 9/2$^-$**	**1902(138)**	**8851(115)**			
^{205}Ac (9/2$^-$)	760(50)	8090(60)						
^{203}Ac 9/2$^-$	**1180(68)**	**8256(38)**						
^{201}Ac 9/2$^-$	**1618(91)**	**8440(51)**						
^{199}Ac 9/2$^-$	**2056(115)**	**8624(64)**						

be determined using the error propagation formula [41], as also presented in Table I. It should be noted that all Q_p values are for the proton emissions from mother nuclei to the 0^+ gs of daughter nuclei, while all Q_α values refer to s-wave α emissions. The reason for making this criterion of $Q_{p/\alpha}$ is that such emissions are the most competitive. For proton emission, these cases correspond to the largest Q_p, and for α decay, they correspond to the smallest centrifugal barrier. Indeed, for the known proton emitter ^{185}Bi and the majority of known α-decaying nuclei in this nuclear region, their decays are dominated by proton emission feeding to the 0^+ gs and s-wave α emission, respectively.

III. Calculation of $T_{1/2}^{p/\alpha}$ values

The estimated Q_p and Q_α values are used to calculate the partial proton-emission and partial α-decay half-lives for these states, respectively. Additionally, the l_p and l_α values are also needed in the calculation. As mentioned in Sec. II, we assume that all α decays are s-wave emission, i.e., l_α=0. Meanwhile, in order to obtain l_p values, the estimation of the possible spin-parities for unknown states based on the known systematics is required.

A. Spin-parity systematics

For the bismuth isotopic chain, the intruder $1/2^+$ configuration in ^{185}Bi becomes the gs, which is in contrast to all odd-A isotopes $^{187-209}$Bi, with the $9/2^-$ gs, see Fig. 5 in Ref. [19]. Based on this, the $1/2^+$ and $9/2^-$ are assumed as the possible spin-parities for the proton-emitting candidates in Bi isotopic chain, as shown in Table I. Similarly, an order reversal of low- and high-spin states occurs in neutron-deficient astatine isotopes, with the $1/2^+$ gs for ^{191,193,195}At while the $9/2^-$ gs are known for heavier isotopes, as shown in Fig. 11 of Ref. [42]. Furthermore, three consecutive low-lying $7/2^-$ states were also observed in ^{191,193,195}At [42]. Therefore, the $1/2^+$ and $7/2^-$ states have been assumed as the possible proton-emitting states in unknown At isotopes.

In the francium isotopic chain, all odd-A isotopes $^{201-213}$Fr have $9/2^-$ gs, while the $1/2^+$ isomers were identified in ^{201,203,205}Fr, see Fig. 12 in Ref. [43]. Based on the rapidly descending trend of the $1/2^+$ levels, the spin-parity of ^{199}Fr gs was tentatively assigned as $(1/2^+)$ [44]. Therefore, the same as in the Bi

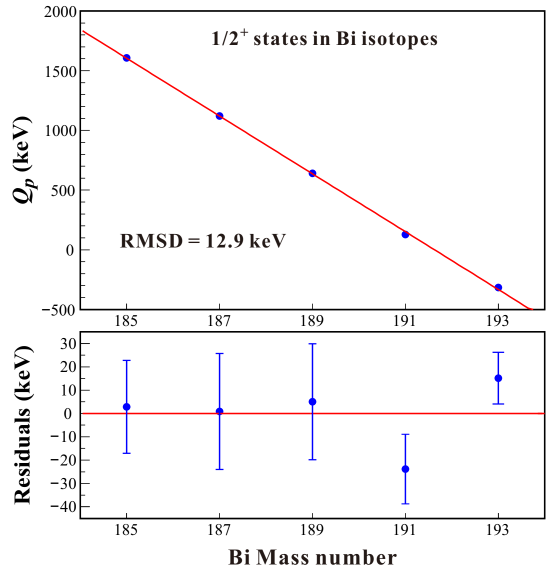

FIG. 2. Upper panel: the Q_p values for the known $1/2^+$ states in odd-A $^{185-193}$Bi isotopes and the corresponding linear fitting. Lower panel: the residuals with uncertainties taken from Q_p values. Data are taken from Refs. [19, 38].

isotopes, both the $1/2^+$ and $9/2^-$ states are considered for Fr isotopes in this study. The gs of ^{197}Fr was tentatively assigned as $(7/2^-)$ according to the systematics of reduced α-decay widths δ^2 [45]. However, no three consecutive $7/2^-$ states were observed in neutron-deficient Fr isotopes.

For the heavier elements actinium and protactinium, the $9/2^-$ gs were identified for all known neutron-deficient isotopes except for ^{203}Ac, where the intruder $1/2^+$ configuration was assumed to become the gs based on the systematics of δ^2 and single-particle energy levels [46]. Moreover, no other given states with three or more consecutive data were observed in neutron-deficient Ac and Fr isotopes [39]. Thus only the $9/2^-$ states are taken into account for the proton-emitting candidates in these two isotopic chains.

B. $T_{1/2}^{p/\alpha}$ calculation in the UDL framework

There are several methods for calculating the partial half-life, as introduced in Sec. I. In this study we choose the UDL approach [28, 29] to calculate $T_{1/2}^{p}$ and $T_{1/2}^{\alpha}$, as it is not only universally valid for all types of charged-particle emissions and for all isotopic series, but also is simple and provides reliable estimates of half-lives. The calculated $T_{1/2}^{p}$ and $T_{1/2}^{\alpha}$ of the specific states in isotopes of interest are shown in Fig. 3. Furthermore, the experimental values are also displayed in Fig. 3 to illustrate the predictive power of the UDL approach.

IV. Results and discussion

Considering the competition between proton emission and α decay, the proton-emitting candidates are defined by the partial half-lives with:

$$T_{1/2}^{p} < 10 T_{1/2}^{\alpha}. \tag{1}$$

This will guarantee that the proton-emission branch ratio is at least 10%. According to the calculated $T_{1/2}^{p}$ and $T_{1/2}^{\alpha}$ shown in Fig. 3 and the criterion (1), we selected proton-emitting candidates, as marked with the brown squares in Fig. 1. Specifically, these candidates are the $1/2^+$ and $9/2^-$ states in ^{183}Bi, the $1/2^+$ and $7/2^-$ states in ^{187,189}At, the $1/2^+$ state in ^{193}Fr and $9/2^-$ state in ^{199}Ac. The calculated $T_{1/2}^{p}$ and $T_{1/2}^{\alpha}$ for these candidates are shown in Table II. It can be noted that the calculated proton-decay half-life of 22^{+8}_{-6} ns for the $1/2^+$ state in ^{183}Bi is consistent with the recent experimental result, which gave an upper limit of 190(60) ns for ^{183}Bi half-life based on its non-observation [47].

TABLE II. The calculated $T_{1/2}^{p}$ and $T_{1/2}^{\alpha}$ for the proton-emitting candidates.

Canditate	$T_{1/2}^{p}$(cal)	$T_{1/2}^{\alpha}$(cal)
^{183}Bi $1/2^+$	22^{+8}_{-6} ns	$2.0^{+0.4}_{-0.3}$ μs
^{183}Bi $9/2^-$	23^{+14}_{-8} μs	210^{+10}_{-10} μs
^{187}At $1/2^+$	200^{+140}_{-80} ns	110^{+30}_{-20} μs
^{187}At $7/2^-$	660^{+750}_{-350} ns	46^{+25}_{-16} μs
^{189}At $1/2^+$	170^{+130}_{-70} μs	660^{+70}_{-60} μs
^{189}At $7/2^-$	510^{+600}_{-270} μs	290^{+60}_{-50} μs
^{193}Fr $1/2^+$	2^{+15}_{-1} ms	140^{+30}_{-20} μs
^{199}Ac $9/2^-$	300^{+1200}_{-200} μs	200^{+100}_{-70} μs

There are two challenges in experimental studies for the predicted proton emitters:

(i) Proton emission is much more sensitive to the decay energy than α decay, causing proton-decay half-lives to become very short rapidly beyond the proton drip line. For instance, in this work the calculated proton-decay half-lives of the $1/2^+$ states in ^{183}Bi and ^{187}At are only 22^{+8}_{-6} ns and 200^{+140}_{-80} ns, respectively. These half-lives are significantly shorter than the typical flight time of $\sim$ 1 μs through currently widely used recoil separator setups, making their observation challenging with existing technology.

(ii) All known proton emitters have mainly been produced via fusion-evaporation reactions. However, as the fusion-fission channel becomes dominant for compound nuclei with $Z > 82$, the cross-sections for synthesizing the most neutron-deficient new isotopes through fusion-evaporation reactions decrease sharply. For example, the production cross-section for the recently synthesized ^{203}Ac by the ^{40}Ca + ^{169}Tm

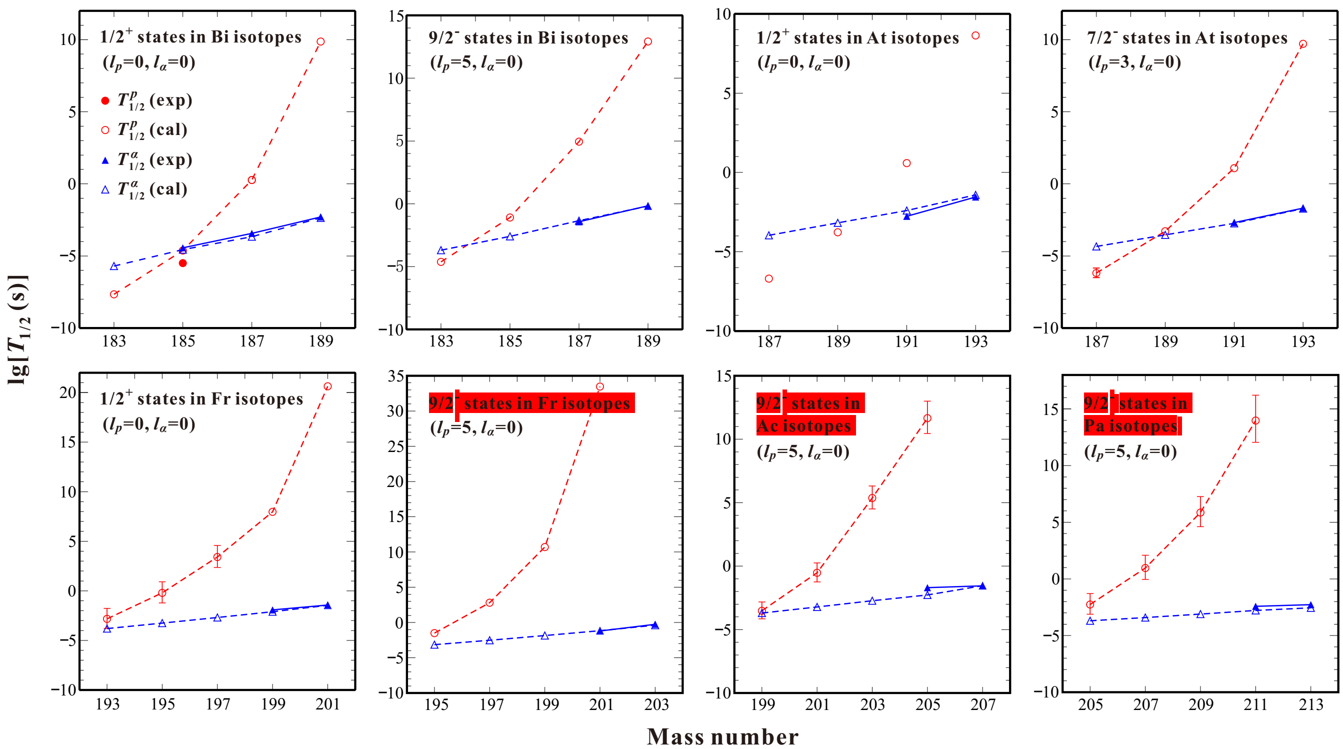

FIG. 3. The partial half-lives for proton emission (circles) and α decay (triangles) of the specific states in the neutron-deficient Bi-Pa isotopes with odd Z and even N. The measured and calculated partial half-lives are shown by solid and open symbols, respectively. The measured half-lives are taken from Ref. [39].

fusion-evaporation reaction is only $0.13^{+0.30}_{-0.10}$ pb [46], which is nearly at the sensitivity limit for the synthesis of new nuclides.

The first factor underscores the necessity of developing advanced detection techniques, e.g., direct detection devices, to identify extremely short-lived proton-emitting candidates near the target. The second factor suggests exploring alternative nuclear-reaction mechanisms besides the fusion-evaporation reaction for synthesizing the heaviest proton emitters, such as projectile-fragmentation reaction [48–51] and multi-nucleon transfer reaction [52, 53].

V. Summary

This study focuses on predicting new proton-emitting candidate states above the Z=82 proton shell by estimating their partial proton-emission and α-decay half-lives. Through a combination of linear extrapolation for deducing $Q_{p/\alpha}$ values and the UDL framework for half-life calculations, eight proton-emission candidate states in five new neutron-deficient isotopes were suggested, namely the $1/2^+$ and $9/2^-$ states in ^{183}Bi, the $1/2^+$ and $7/2^-$ states in ^{187,189}At, the $1/2^+$ state in ^{193}Fr and $9/2^-$ state in ^{199}Ac. Furthermore, this work highlights the importance of developing new detection techniques and exploring alternative nuclear-reaction mechanisms besides the fusion-evaporation reaction to search for the heaviest proton emitters.

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