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New α-Emitting Isotope ^{214}U and Abnormal Enhancement of α-Particle Clustering in Lightest Uranium Isotopes

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A new α-emitting isotope ^{214}U, produced by the fusion-evaporation reaction ^{182}W$(^{36}$Ar, 4n$)^{214}$U, was identified by employing the gas-filled recoil separator SHANS and the recoil-α correlation technique. More precise α-decay properties of even-even nuclei ^{216,218}U were also measured in the reactions of ^{40}Ar, ^{40}Ca beams with ^{180,182,184}W targets. By combining the experimental data, improved α-decay reduced widths δ^2 for the even-even Po-Pu nuclei in the vicinity of the magic neutron number $N = 126$ are deduced. Their systematic trends are discussed in terms of the $N_p N_n$ scheme in order to study the influence of proton-neutron interaction on α decay in this region of nuclei. It is strikingly found that the reduced widths of ^{214,216}U are significantly enhanced by a factor of two as compared with the $N_p N_n$ systematics for the $84 \leq Z \leq 90$ and $N < 126$ even-even nuclei. The abnormal enhancement is interpreted by the strong monopole interaction between the valence protons and neutrons occupying the $\pi 1f_{7/2}$ and $\nu 1f_{5/2}$ spin-orbit partner orbits, which is supported by the large-scale shell model calculation.

Nucleon-nucleon interaction, which governs the existence of nuclear system, plays a fundamental role in understanding the properties of exotic nuclei far from stability. Although the proton-proton (p-p) and neutron-neutron (n-n) correlations are well known to be crucial for explaining a wealth of experimental data, the proton-neutron (p-n) interaction has long been recognized as one of the essential driving forces for the shell structure evolution, the development of collectivity, and the onset of deformation in atomic nuclei [1–10]. In the last decades, thanks to the development of radioactive beam facilities worldwide, enormous progress in the physics of change of nuclear shell structure as a function of proton/neutron numbers has been achieved in light nuclei. However, when going from lead towards uranium isotopes, the experimental knowledge for the structure evolution in the proximity of the closed shell at $N = 126$ becomes increasingly scarce [3,11–15].

It is well known that, because of large overlap of the radial wave functions, the attractive and short-range interaction between valence protons and neutrons occupying orbits with the same number of nodes and orbital angular momenta (i.e., $\Delta n = \Delta l = 0$) becomes stronger than those in other categories, and eventually triggers the remarkable changes of closed shells (see [3,4] and references therein). For instance, the $\pi 0 f_{7/2} - \nu 0 f_{5/2}$ interaction was shown to play an important role in the structure evolution of $N = 34$ isotones of Ca, Ti, Cr, and Fe, culminating in the creation of new magic numbers at $N = 32, 34$ in ^{52,54}Ca (see Fig. 1 from Ref. [16] and Ref. [17]). In the translead nuclear region with $Z > 82$ and $N \leq 126$, the valence protons fill the $0h_{9/2}$, $1f_{7/2}$, and $0i_{13/2}$ orbits, while the neutrons mainly occupy the $2p_{1/2}$, $1f_{5/2}$, and $2p_{3/2}$ orbits [18,19]. Therefore, one can expect the monopole p-n interaction between the $\pi 1 f_{7/2}$ and $\nu 1 f_{5/2}$ spin-orbit partner orbits to have a significant impact on nuclear structure evolution in that region.
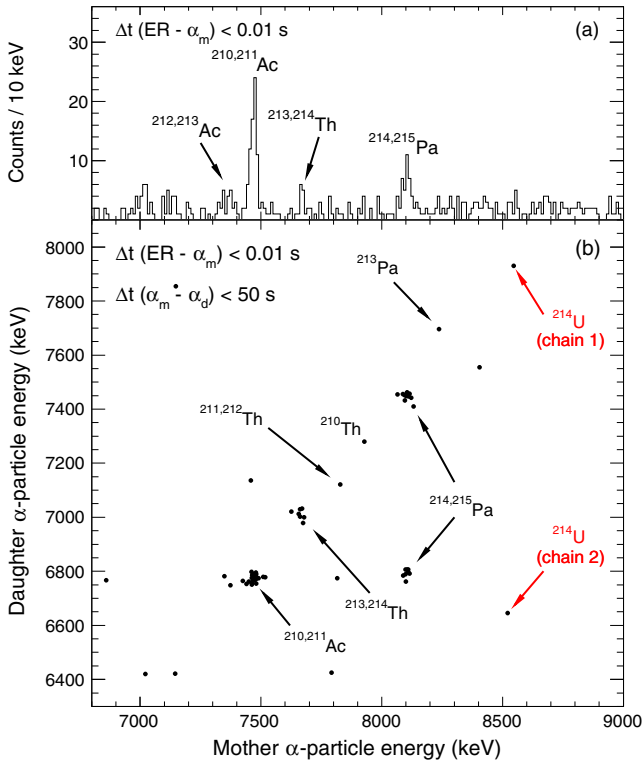

FIG. 1. (a) Energy spectrum for α-decay events following recoil implantations within a time window of 10 ms. (b) Two-dimensional plot of mother and daughter α-particle energies for $ER - \alpha_m - \alpha_d$ correlations in the ^{36}Ar + ^{182}W reaction. Maximum search times for the $ER - \alpha_m$ and $\alpha_m - \alpha_d$ pairs are 10 ms and 50 s, respectively. The decay events from the new isotope ^{214}U labeled as "chain 1" and "chain 2" are indicated by red arrows. The ^{210}Th event is from the decay of daughter nucleus of ^{214}U in chain 1. The details of these decay chains can be found in Fig. 2.

α-decay spectroscopy has been proven to be a powerful tool to probe the nuclear structure in heavy nuclei [20–23]. There are analytical formulae to calculate the α-decay half-lives such as the new Geiger-Nuttall law [24,25]. Typically, the α-decay process is described by the two-step mechanism, involving the preformation of an α particle followed by its penetration through Coulomb and centrifugal barriers. The α-particle preformation probability involves all the nuclear structure information and can be weighed experimentally by the α-decay reduced width δ^2 [26] or the model-independent formation probability $|R\mathcal{F}_\alpha(R)|^2$ [27,28]. It is interesting to note that the α-decay reduced widths of several $Z \sim N$ nuclei around ^{100}Sn ($Z = N = 50$) are enhanced by at least a factor of two relative to the benchmark nucleus $^{212}_{84}$Po$_{128}$ and its neighboring Po isotopes [29–32]. This enhancement was explained by the so-called superallowed α decay [20,33] in relation to the fact that the valence protons and neutrons are in the same single-particle levels, giving rise to the strong p-n interaction. In fact, the influence of p-n interaction on the absolute α-decay widths in ^{212}Po and nearby nuclei was usually neglected in microscopic calculation, since the low-lying proton and neutron single-particle states are very different from each other in these cases [21,34,35]. However, several theoretical treatments [36–38] pointed out the particular significance of p-n interaction in α decay for these nuclei.

In this Letter, we report on the observation of a new isotope $^{214}_{92}$U$_{122}$ and on more precise measurements of the α-decay properties of ^{216,218}U ($N = 124, 126$). In this region of nuclei, the protons and neutrons can occupy the $\pi 1 f_{7/2}$ and $\nu 1 f_{5/2}$ spin-orbit partner orbits to a large extent. Thus, such nuclei can provide a unique opportunity to test the influence of p-n interaction on the α-particle clustering in heavy nuclear region.

To produce ^{214,216,218}U nuclei, a series of experiments were performed at the gas-filled recoil separator, SHANS (Spectrometer for Heavy Atoms and Nuclear Structure) [39], at the Heavy Ion Research Facility in Lanzhou (HIRFL), China. For ^{214}U, the fusion-evaporation reaction of ^{182}W(^{36}Ar, 4n)^{214}U with a beam energy of 184 MeV and a typical beam intensity of ~500 pnA was used. The ^{182}W targets with a thickness of 300–350 μg/cm^2 were prepared by sputtering the material onto 80-μg/cm^2-thick carbon foils and then covered by 10-μg/cm^2-thick carbon layer. The recoiled evaporation residues (ERs) were separated efficiently by SHANS and collected by three 16-strip position-sensitive silicon detectors (PSSDs), which were mounted side by side at the focal plane of the separator. Each PSSD has an active area of 50×50 mm^2. Due to the shallow implantation depth of ~4 μm, the full-energy α particles emitted from ERs and/or their descendants were registered with an efficiency of ~54%. The typical energy resolution for the PSSDs was 35 keV (FWHM) for 6–9 MeV α particles. Eight side silicon detectors (SSDs)

were mounted in front of the PSSDs in an open box geometry to measure the escaped α particles with an extra detection efficiency of ~18%. For such events, the total α-particle energy was reconstructed by adding the deposited energies in PSSD and SSD. In order to distinguish the α-decay events from the implanted products, two multiwire proportional counters were installed upstream from the PSSDs. A digital data readout system including waveform digitizers was used for the data acquisition. Details of the detection system and data analysis method were described in Refs. [14,15,40,41].

The identification of ^{214}U was performed by searching for the position-time correlated α-decay chains with the help of known α-decay properties of its descendants. An energy spectrum for the α-decay events following the ERs and a two-dimensional plot for the decay energy correlation between mother and daughter nuclei (ER $- \alpha_m - \alpha_d$) are shown in Figs. 1(a) and 1(b), respectively. The Pa, Th, and Ac isotopes were produced from charged-particle evaporation channels. Two decay events in Fig. 1(b) were assigned to the new isotope ^{214}U unambiguously. The details of these decay chains are displayed in Fig. 2. The measured decay properties of daughter products match well with the known data [42] for ^{210}Th, ^{206}Ra, ^{202}Rn, and ^{198}Po. Based on these measurements, the mean α-particle energy and half-life of ^{214}U were determined to be 8533(18) keV and $0.52^{+0.95}_{-0.21}$ ms, respectively, which are listed in Table I. The uncertainties of half-life were estimated by the maximum likelihood method described in Ref. [43]. The production cross section for ^{214}U was determined to be 10^{+14}_{-7} pb.

The properties of ^{216}U, which was the lightest even-even uranium isotope known previously, were reported in our previous work [44] and in Refs. [45,46]. However, at most four decay chains from the ground state of ^{216}U were observed in each study, resulting in a relatively large uncertainty of decay half-life. In the present investigation,
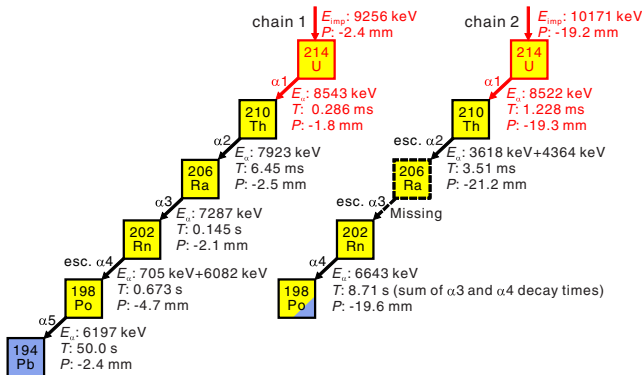
the same experimental setup as for ^{214}U was used, but with a reaction of ^{180}W(^{40}Ar, 4n)^{216}U at a beam energy of 191 MeV. Thirteen decay chains were assigned to the ground-state-to-ground-state (g.s.-to-g.s.) decay of ^{216}U. The deduced decay energy and half-life of ^{216g}U are 8374(17) keV and $1.28^{+0.49}_{-0.28}$ ms, respectively. By combining all the data from the present study and from Refs. [44–46], the averaged half-life for the ground state of ^{216}U was deduced to be $2.25^{+0.63}_{-0.40}$ ms. The results are compared with the literature data in Table I.

In order to obtain more precise decay properties of ^{218}U, two experiments with the ^{182}W(^{40}Ar, 4n)^{218}U and ^{184}W(^{40}Ca, α2n)^{218}U reactions were carried out with beam energies of 190 and 206 MeV, respectively. In total, 76 decay chains were assigned to the decay from the ground state of ^{218}U, leading to the determination of $E_\alpha = 8612(14)$ keV and $T_{1/2} = 0.65^{+0.08}_{-0.07}$ ms. The uncertainties of half-life were improved in comparison with previous results [44,47–49] (see Table I).

To study the nuclear structure evolution in the $N = 126$ region, the reduced widths δ^2 for g.s.-to-g.s. decays of even-even $84 \leq Z \leq 94$ nuclei are extracted by using Rasmussen method [26], see Fig. 3(a). The uncertainties of δ^2 values are mostly due to the half-life uncertainties. The ^{214,216,218}U values determined in this work are shown in column 4 of Table I and plotted by filled circles in Fig. 3(a).

In each of the Po, Rn, Ra, and Th isotopic chains, a sharp decrease of reduced widths at $N = 126$ is well established, indicating a notable neutron shell effect [26,54,55]. Our new data suggest for the first time that the minimum decay width for U isotopes is likely at ^{218}U ($N = 126$). This result is in contrast with our previous work [44], where a lower value of $\delta^2(^{216}$U$) = 34^{+34}_{-11}$ keV was reported. A shrinking of the δ^2 enhancement between the $N = 126$ and $N = 130$ isotones with the increasing proton number was attributed

FIG. 2. Observed α-decay chains for ^{214}U. For each chain, the implantation energy of ERs (E_{imp}), the α-particle energy (E_α), the decay time (T), and the position (P) in the strip detector are shown. The reconstructed energies for escaping α decays are given as the sum of the PSSD and SSD energies.

TABLE I. The g.s.-to-g.s. α-decay energies and half-lives of ^{214,216,218}U measured in this work. The reduced α-decay widths δ^2, in column 4, are calculated by Rasmussen formalism [26] assuming the α-particle angular momentum, $\Delta L = 0$. The data for ^{216,218}U are compared with literature values.

	This work			Literature data		
Isotope	E_α/keV	$T_{1/2}$/ms	δ^2/keV	E_α/keV	$T_{1/2}$/ms	Ref.
^{214}U	8533(18)	$0.52^{+0.95}_{-0.21}$	128^{+233}_{-52}	$\cdots$	$\cdots$	$\cdots$
^{216}U	8374(17)	$2.25^{+0.63a}_{-0.40}$	78^{+22}_{-14}	8384(30)	$4.72^{+4.72}_{-1.57}$	[44]
				8340(50)	$3.8^{+8.8}_{-3.2}$	[45]
				8390(33)	$2.6^{+3.6}_{-1.0}$	[46]
^{218}U	8612(14)	$0.65^{+0.08}_{-0.07}$	53^{+7}_{-6}	8600(30)	$1.15^{+1.58}_{-0.42}$	[44]
				8612(9)	$0.51^{+0.17}_{-0.10}$	[47,48]
				8625(25)	$1.5^{+7.3}_{-0.7}$	[49]

^{a}The value is deduced by combining all 21 decay events from this work and Refs. [44–46], and is also used for the decay width calculation for ^{216}U.
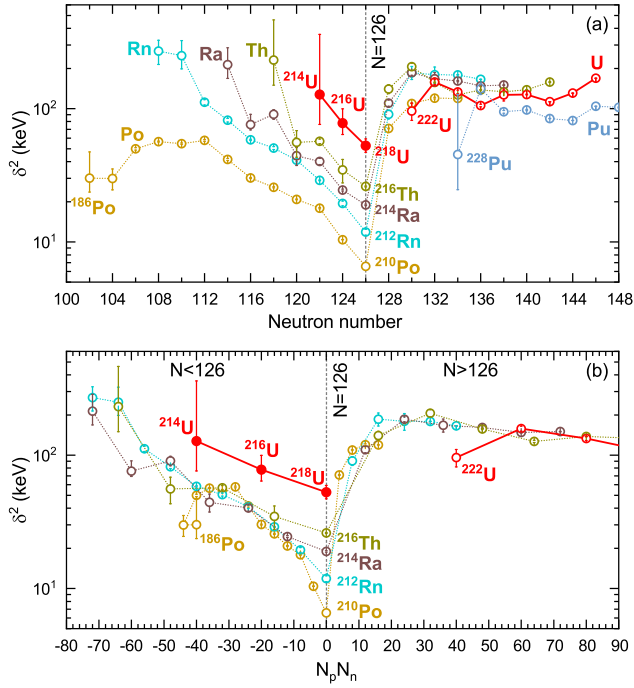
FIG. 3. (a) Systematics of reduced widths for g.s.-to-g.s. α decays of even-even $84 \leq Z \leq 94$ isotopes as a function of neutron number. The decay properties are taken from Refs. [11,12,42,50–53]. The values for ^{214,216,218}U from this work are shown by filled circles. The errors of reduced widths are only determined by half-life uncertainties. (b) Same as (a) but against $N_p N_n$ for even-even Po to U isotopes. The N_p and N_n values are calculated relative to $Z = 82$ and $N = 126$ closed shells, respectively, with an exception of $^{186}_{84}$Po$_{102}$, for which $N_n = -20$, relative to the closest $N = 82$ neutron shell.

to the weakening of the $N = 126$ shell effect as suggested in Ref. [12]. The nearly constant or even decreasing values for the most neutron-deficient polonium isotopes were explained by the configuration mixing effect [11,20].

In Fig. 3(a), another important feature revealed by our new data is that, while the decay widths at $N = 122, 124$, and 126 for Po-Th isotopes increase monotonously with the increasing proton number, an unexpected sharp increase was observed from Th to U isotopes at the same neutron numbers. This suggests that the α-particle formation probability is enhanced in these U isotopes.

In order to get a deeper insight into the behavior of reduced widths, we studied the influence of the p-n interaction upon the α-decay process in this mass region. Given the fact that the $N_p N_n$ scheme [56,57] allows a uniform description of structure evolution for a variety of observables and highlights the importance of valence p-n interaction [1,2,58–61], the δ^2 values are plotted against $N_p N_n$ in Fig. 3(b). Here, N_p and N_n are the numbers of valence protons and neutrons relative to the nearest closed shells: $Z = 82$ for proton and $N = 126$ for neutron. It is striking to see that the $N_p N_n$ plot displays a remarkable simplification for the systematics of decay widths in this

region. In the $N > 126$ region, the δ^2 values increase rapidly until $N_p N_n \approx 20$, and then converge into a nearly constant value of ~ 150 keV (except for ^{222}U). This "saturation" phenomenon might indicate that the α decays in these nuclei are affected only slightly by the p-n interaction, but are dominated by the p-p and n-n pairing interactions, as pointed out theoretically in Refs. [21,34,35]. In other words, it is the strong pairing force among the protons and neutrons occupying high-j orbits (e.g., $\pi 0h_{9/2}$ and $\nu 1g_{9/2}$), which leads to the large α-particle formation probability [11].

In contrast, for the $N < 126$ nuclei, the δ^2 values for Po-Th isotopes show quite different behaviors, increasing exponentially with increasing the absolute $N_p N_n$ quantity along a relatively compact tendency (except for ^{186,188}Po). This increasing trend can be partly explained by the increasing neutron and proton pairing correlations [11] as one moves away from the $N = 126$ and $Z = 82$ shells. More importantly, considering that the $N_p N_n$ value provides a reliable measure of interaction between the valance protons and neutrons [1,2], this specific feature shown in Fig. 3(b) implies that the p-n interaction can also play an essential role in the α-particle clustering in this region.

The δ^2 values of ^{214,216}U, however, show striking discrepancy with the unified trend established for $84 \leq Z \leq 90$ and $N < 126$ nuclei. Regardless the relatively large uncertainties for ^{214}U, a significant enhancement by a factor of two is revealed for ^{214,216}U as shown in Fig. 3(b). This new feature might be related to the possible changes of occupancy of $0h_{9/2}$ and $1f_{7/2}$ proton orbits approaching $Z = 92$. Indeed, below $Z = 92$, it is expected that the $0h_{9/2}$ protons play a dominant role, which is confirmed by, e.g., the $9/2^-$ ground states for most of odd-A $_{83}$Bi, $_{85}$At, $_{87}$Fr, and $_{89}$Ac isotopes [42]. The $0h_{9/2}$ orbit is expected to be highly occupied in U ($Z = 92$) with an enhanced probability of proton occupancy of the higher-lying $1f_{7/2}$ orbit. The latter, combined with the neutron occupancy of $1f_{5/2}$ orbit around $N = 118$–124, might lead to a strong monopole p-n interaction [see inset of Fig. 4(b)], which enhances the preformation probability in α decay.

In order to verify this conjecture, we have performed large-scale shell model calculations for the $84 \leq Z \leq 94$ and $N = 122, 124, 126$ even-even nuclei. Taken ^{208}Pb as a core, the valence protons and hole neutrons are considered for these nuclei. The $0h_{9/2}$, $1f_{7/2}$, and $0i_{13/2}$ orbits beyond the $Z = 82$ shell are used for protons, while the $2p_{1/2}$, $1f_{5/2}$, and $2p_{3/2}$ orbits below the $N = 126$ shell are used for neutrons. The single-particle energies are fixed to those of ^{209}Bi and ^{207}Pb. The p-p, n-n, and p-n parts of two-body interactions are taken from the Kuo-Herling particle interaction [62], Kuo-Herling hole interaction [63], and monopole based universal interaction [64] plus M3Y spin-orbit interaction [65], respectively. The calculated proton occupation numbers for the $\pi 0h_{9/2}$, $\pi 1f_{7/2}$, and $\pi 0i_{13/2}$ orbits in
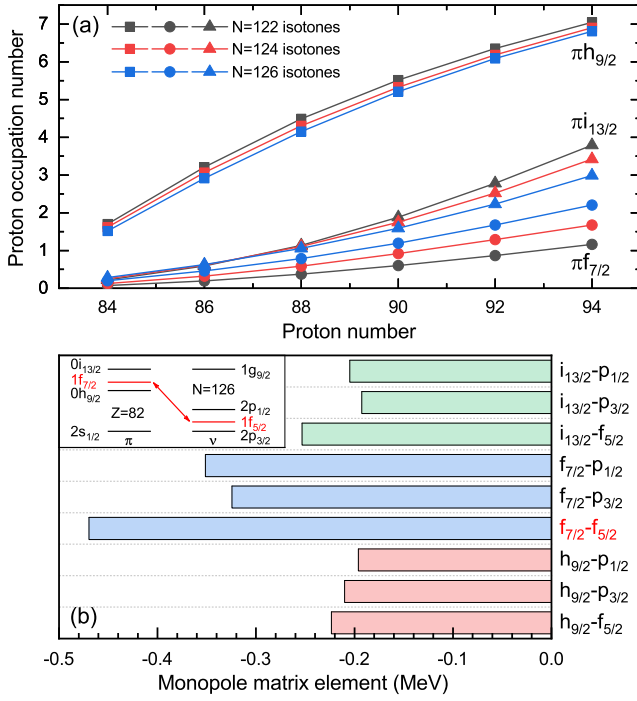
FIG. 4. (a) Calculated proton occupation numbers for the $\pi 0h_{9/2}$ (square), $\pi 1f_{7/2}$ (circle), and $\pi 0i_{13/2}$ (triangle) orbits in $N = 122$, 124, and 126 even-Z isotones of Po-Pu. (b) Monopole matrix elements of p-n interaction calculated for the $Z > 82$ and $N \leq 126$ nuclei. The inset in panel (b) shows the single-particle orbits near the ^{208}Pb doubly closed shells, and the strong interaction between the $1f_{7/2}$ protons and $1f_{5/2}$ neutrons is marked.

$N = 122$, 124, and 126 even-Z isotones are shown in Fig. 4(a). It can be seen that, due to the pairing correlation effect [1], the valence protons occupy mainly the $0h_{9/2}$ orbit with the increasing occupation probability of the $1f_{7/2}$ and $0i_{13/2}$ protons from Po to Pu isotopes. In particular, the effective proton occupation numbers for the $1f_{7/2}$ orbit in U and Pu isotopes are almost equal to or even higher than one.

The calculated monopole matrix elements between the proton and neutron orbits for the $Z > 82$ and $N \leq 126$ nuclei are shown in Fig. 4(b). The calculations demonstrate that all the p-n interactions involving $1f_{7/2}$ protons are about twice more attractive than those involving $0h_{9/2}$ and $0i_{13/2}$ protons. In particular, the $\pi 1f_{7/2} - \nu 1f_{5/2}$ interaction is by far the strongest one in this region of nuclei. Therefore, the strong p-n interactions related to the $1f_{7/2}$ protons, together with the increased occupancy of the $\pi 1f_{7/2}$ orbit, would lead to the enhanced α-particle formation probability in the $N = 122$, 124, and 126 uranium isotopes.

In summary, a new isotope ^{214}U was identified and improved α-decay properties of ^{216,218}U were measured by employing the gas-filled recoil separator SHANS and the recoil-α correlation method. By combining the new and

previously known data, we extracted the α-decay reduced widths δ^2 for the even-even Po–Pu nuclei with Rasmussen method. It is found that the δ^2 systematics from Po to Th can be merged into two compact trends for the $N < 126$ and $N > 126$ nuclei in terms of the $N_p N_n$ scheme. The behavior in the $N < 126$ region indicates a crucial role played by the p-n interaction in α decay. Meanwhile, it is strikingly found that the reduced widths of ^{214,216}U are enhanced remarkably by a factor of two relative to the systematic trend for the $N < 126$ nuclei in the $N_p N_n$ scheme. This might be explained as being due to the strong monopole interaction between the valence $1f_{7/2}$ protons and $1f_{5/2}$ neutrons combined with the increased occupancy of the $f_{7/2}$ proton orbit, which was confirmed by the large-scale shell model calculations.

As a possible outlook for the future studies in this region, it is expected that, in view of the continuously increasing proton occupancy of the $1f_{7/2}$ orbit and the further enhancement of p-n interaction, this effect might become even stronger in the Pu isotopes. Thus, it is extremely intriguing to extend the δ^2 systematics to higher-Z nuclei.

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