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 fusion-evaporation reaction ^{182}W(^{36}Ar, 4n)^{214}U, was identified by employing the gas-filled recoil separator SHANS and recoil-α correlation technique. More precise α-decay properties of even-even nuclei ^{216,218}U were also measured in reactions of ^{40}Ar, ^{40}Ca 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 magic neutron number $N = 126$ were deduced. Their systematic trends are discussed in terms of N_pN_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_pN_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 a large-scale shell model calculation.

Nucleon-nucleon interaction, which governs the existence of nuclear system, plays a fundamental role in understanding of 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 and/or neutron numbers has been achieved in light nuclei. However, the experimental knowledge for the structure evolution in heavy nuclei around and below the neutron closed shell at $N = 126$ remains scarce at present [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 $\pi 0f_{7/2}$–$\nu 0f_{5/2}$ interaction was shown to play an important role for 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 trans-lead 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 $\pi 1f_{7/2}$ and $\nu 1f_{5/2}$ spin-orbit partner orbits to have a significant impact on nuclear structure evolution in that region.

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 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 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 neighbouring 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 a 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 to 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 for the α-decay properties of ^{216,218}U ($N = 124, 126$). In this region of nuclei, the protons and neutrons can occupy the $\pi 1f_{7/2}$ and $\nu 1f_{5/2}$ 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 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 $\sim$500 pnA was used. The ^{182}W targets with a thickness of 300-350 μg/cm^2 were prepared by sputtering the material onto 80-μg/cm^2-thick carbon foils and then covered by 10-μg/cm^2-thick carbon layer. The recoiled evaporation residues (ERs) were separated efficiently by SHANS and collected by three 16-strip position-sensitive silicon detectors (PSSDs), which were mounted side by side at the focal plane of the separator. Each PSSD has an active area of 50$\times$50 mm^2. Due to the shallow implantation depth of $\sim$4 μm, the full-energy α particles emitted from ERs and/or their descendants were registered with an efficiency of $\sim$54%. Eight side silicon detectors (SSDs) were mounted in front of the PSSDs in an open box geometry to measure the escaped α particles. 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 multi-wire 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 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 Fig. 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

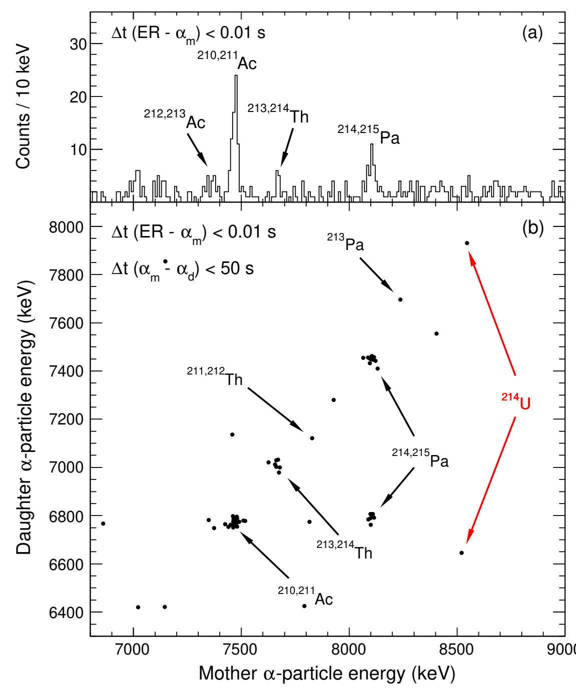

FIG. 1. a) Energy spectrum for α-decay events following recoil implantations within a time window of 10 ms. b) Two-dimensional plot of mother and daughter α-particle energies for ER $- \alpha_m - \alpha_d$ correlations in the ^{36}Ar+^{182}W reaction. Maximum search times for the ER $- \alpha_m$ and $\alpha_m - \alpha_d$ pairs are 10 ms and 50 s, respectively. The decay events from the new isotope ^{214}U are indicated by red arrows.

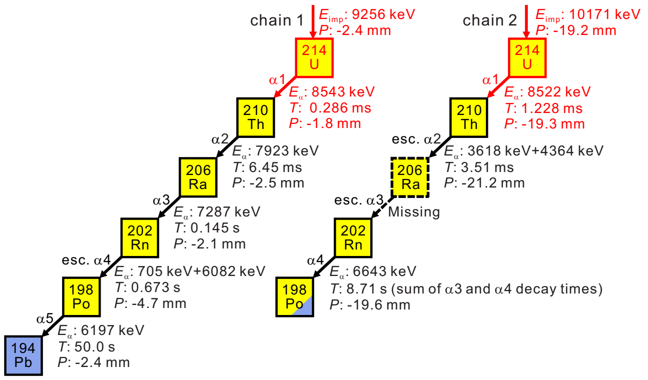

FIG. 2. Observed α-decay chains for ^{214}U. For each chain, the implantation energy of ERs ($E_{\rm imp}$), the α-particle energy (E_α), the decay time (T), and the position (P) in the strip detector are shown. The reconstructed energies for escaping α decays are given as the sum of the PSSD and SSD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

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 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 ^{182}W(^{40}Ar, 4n)^{218}U and ^{184}W(^{40}Ca, α2n)^{218}U reactions were carried out with beam energies of 190 MeV and 206 MeV, respectively. Totally, 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 only half a value of $\delta^2(^{216}\mathrm{U})$ (34^{+34}_{-11} keV) was reported. A shrinking of the δ^2 enhancement between the $N = 126$ and $N = 130$ isotones with the increasing of proton number was attributed to a 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 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_pN_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_pN_n in Fig. 3(b). Here, N_p and N_n are the

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.

Isotope	This work			Literature data		
	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}	-	-	-
				8384(30)	$4.72^{+4.72}_{-1.57}$	[44]
^{216}U	8374(17)	$2.25^{+0.63}_{-0.40}$ [a]	78^{+22}_{-14}	8340(50)	$3.8^{+8.8}_{-3.2}$	[45]
				8390(33)	$2.6^{+3.6}_{-1.0}$	[46]
				8600(30)	$1.15^{+1.58}_{-0.42}$	[44]
^{218}U	8612(14)	$0.65^{+0.08}_{-0.07}$	53^{+7}_{-6}	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.

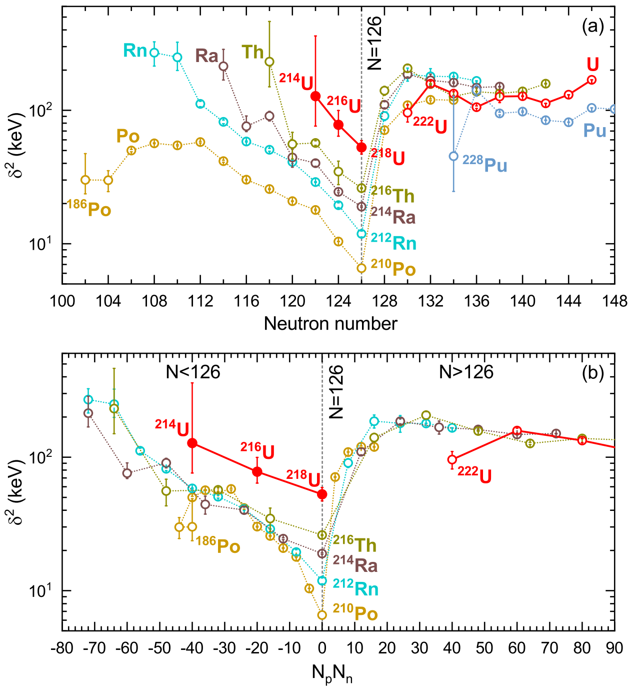

FIG. 3. (a) Systematics of reduced widths for g.s.-to-g.s. α decays of even-even $84 \leq Z \leq 94$ isotopes as a function of neutron number. The decay properties are taken from Refs. [11, 12, 42, 50–53]. The values for ^{214,216,218}U from this work are shown by filled circles. The errors of reduced widths are only determined by half-life uncertainties. (b) Same as (a) but against N_pN_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.

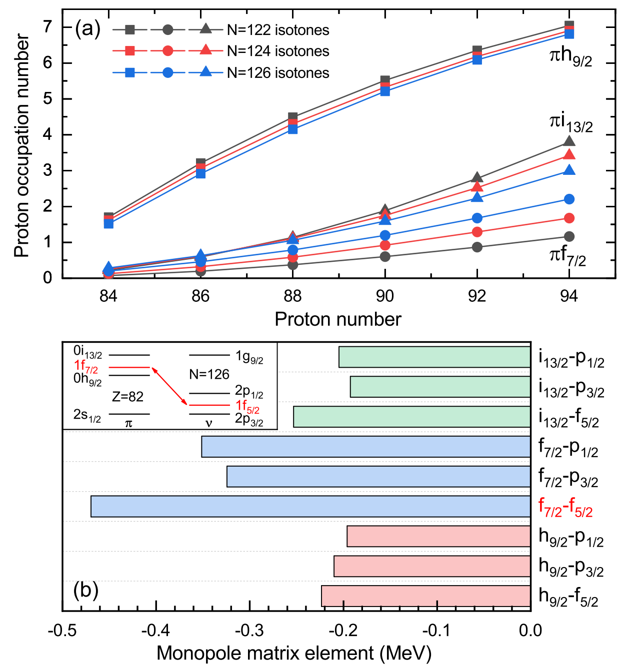

FIG. 4. (a) Calculated proton occupation numbers for the $\pi 0h_{9/2}$ (square), $\pi 1f_{7/2}$ (circle), and $\pi 0i_{13/2}$ (triangle) orbits in $N = 122$, 124, and 126 even-Z isotones of Po–Pu. (b) Monopole matrix elements of p-n interaction calculated for the $Z > 82$ and $N \leq 126$ nuclei. The 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.

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_pN_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 $N_pN_n \approx 20$, 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_pN_n quantity along a relatively compact tendency (except for ^{186,188}Po). The increasing trend can be partly explained by the increasing neutron and proton pairing correlations [11] as one moves away from $N = 126$ and $Z = 82$. More importantly, considering that the N_pN_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 later, 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. The same model spaces with the $0h_{9/2}$, $1f_{7/2}$, $0i_{13/2}$, $2p_{3/2}$, $1f_{5/2}$, and $2p_{1/2}$ orbits were selected for protons and 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. Given the computational limit, the restrictions, for which the $\pi(2p_{3/2},\ 1f_{5/2},\ 2p_{1/2})$ orbits are fully empty for protons and the $\nu(0h_{9/2},\ 1f_{7/2},\ 0i_{13/2})$ orbits are fully occupied for neutrons, were made. The calculated proton occupation numbers for the $\pi 0h_{9/2}$, $\pi 1f_{7/2}$, and $\pi 0i_{13/2}$ orbits in $N = 122$, 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 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 N_pN_n scheme. The behavior in the $N < 126$ region indicates a crucial role played by *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 of $N < 126$ nuclei in the N_pN_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 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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