Letter

New isotope ^{207}Th and odd-even staggering in α-decay energies for nuclei with $Z > 82$ and $N < 126$

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The new thorium isotope ^{207}Th has been produced in the 5n evaporation channel of the fusion reaction ^{36}Ar + ^{176}Hf. It was separated in flight by the gas-filled recoil separator SHANS and identified on the basis of a correlated α-decay chain. The α decay of ^{207}Th, measured with an α-particle energy of 8167(21) keV and a half-life of $9.7^{+46.6}_{-4.4}$ ms, is assigned to originate from ground state. By combining with existing data, we find that the α-decay energies of nuclei with $Z > 82$ and $N < 126$ show a regular and distinct odd-even staggering (OES) rather than the commonly supposed smooth pattern. A theoretical analysis has been performed within relativistic Hartree-Fock-Bogoliubov and large-scale shell-model approaches. It is found that the OES originates from both pairing correlations and blocking of particular orbitals by unpaired nucleons. Of particular importance is that pairing correlations result in the OES not only through the contribution of pairing energy to binding energy, but also by configuration mixing induced by scattering nucleons to orbitals away from Fermi levels.

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Atomic nuclei are quantum many-body systems composed of protons and neutrons. Many quantities describing nuclear properties, such as nuclear binding energy (or mass) [1], one-nucleon separation energy [2–4] and nuclear charge radius [5–8], display odd-even staggering (OES) while changing proton or neutron number. This phenomenon is generally ascribed to the nucleon pairing correlations which are well incorporated in contemporary nuclear theories [9–11]. Relying on the extensive studies on α-decay process, the OES was also found in α-preformation probabilities and α-decay half-lives [12–15]. The OES in α-preformation probabilities, i.e., an odd-$N(Z)$ nuclide has a smaller α-preformation probability than that of its even-$N(Z)$ neighbors, is attributed to the blocking effect of unpaired nucleons [13,14]. Considering that there is no obvious odd-even difference in the penetration process, the OES in α-decay half-lives is proposed to be mainly related with the α-preformation probabilities [15]. However, based on the classical Bethe-Weizsäcker mass formula, one usually believes that there is no or little OES in α-decay energies (Q_α) owing to the fact that a parent nucleus and its daughter share the same odevity of nucleon numbers, and the pairing terms in the binding energies of parent and daughter nuclei could be canceled to a large extent leading to a negligible contribution of pairing energy to the Q_α value [16–18].

In the region of $Z > 82$ and $N < 126$, nuclei decay prevailingly by emitting α particles, and extensive experimental studies provide systematic α-decay data. The Q_α values are generally measured with a high precision of about 20 keV. Even in the case of very low statistics, the achieved precision is still comparable to those from the state-of-the-art mass measurements. We have studied the α-decay properties of ^{205}Ac [19] and $^{214-216}$U [20–22] in this region. As a consequence, we find that the Q_α values along an isotopic chain follow actually a stepwise rather than smooth upward trend with decreasing neutron number, and surprisingly the odd-even feature is even visible to the naked eyes. This finding is obviously at odds with the thought mentioned above, which motivates us to continue α-decay studies in this region and particularly to investigate the OES in α-decay energies.

In this Letter, we report the discovery of the isotope ^{207}Th. By combining the newly measured α-decay properties with

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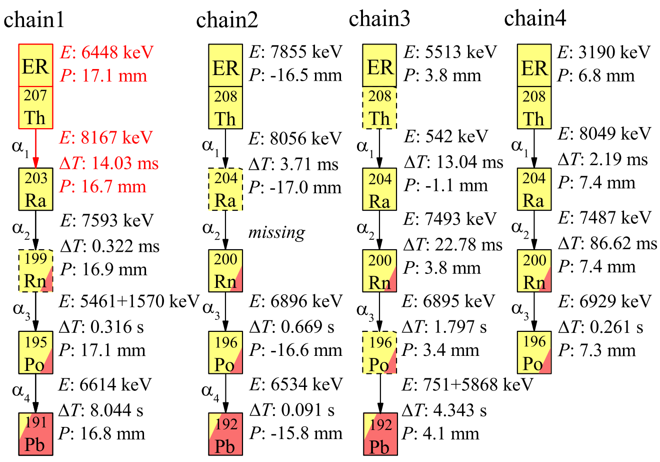

FIG. 1. The observed α-decay chains assigned to ^{207}Th and ^{208}Th. The annotations are the measured energy (E), decay time (ΔT), and vertical position (P) for each event within the chains. Escaped α decays are marked by rectangles with dashed frames.

existing data, we obtain direct evidence that there is a regular and distinct OES in α-decay energies for nuclei with $Z > 82$ and $N < 126$ along both isotopic and isotonic chains. The mechanisms of the OES are discussed on the basis of the relativistic Hartree-Fock-Bogoliubov (RHFB) [23–25] and large-scale shell-model (LSSM) [26,27] calculations.

The experiment to produce ^{207}Th was performed using the ^{176}Hf(^{36}Ar, $5n$) ^{207}Th reaction at the Spectrometer for Heavy Atoms and Nuclear Structure (SHANS) [28]. The 197–199 MeV ^{36}Ar^{11+} beam with a typical intensity of $\sim$0.4 pμA was delivered by the Sector Focusing Cyclotron of the Heavy Ion Research Facility in Lanzhou (HIRFL), China. Isotopically enriched (84.6%) ^{176}Hf targets with thicknesses of 116–360 μg/cm^2 were mounted on a rocking frame which moves horizontally and periodically from side to side during irradiation. The evaporation residues (ERs) were collected and separated by the SHANS and then implanted into three 300-μm-thick position-sensitive silicon strip detectors (PSSDs) surrounded by eight non-position-sensitive silicon detectors (side detectors). All silicon detectors were cooled to a temperature of 251 K using circulating alcohol. Energy resolutions (full width at half maximum) of individual strips of the PSSDs were about 35 keV for 6–10-MeV α particles, and vertical position resolutions are better than 1.2 mm. In the case of escaped α particles deposited less than 2-MeV energy in the PSSDs, the position resolutions were deteriorated to be 3–5 mm. In order to distinguish the radioactive decay events from the implantation events, two multiwire proportional counters were mounted in front of the PSSDs. For details on experimental setup and data analysis see our previous papers [28,29].

To identify the nuclei of interest, a search for decay chains with two to four consecutive α decays, starting from the recoil implantation was performed. As shown in Fig. 1, an α-decay chain (chain1) assigned to the new isotope ^{207}Th was found. The probability of random correlations [30], calculated on the basis of average counting rates in the detectors, was estimated to be less than 1.0×10^{-12}. This indicates that this chain is very unlikely due to random correlations of unrelated events. The measured energy and decay time of the α_2 event are 7593 keV and 0.322 ms, respectively. It can be recognized as belonging to the known isotope ^{203}Ra whose reported α-decay properties are $E_\alpha = 7589(8)$ keV, $T_{1/2} = 31^{+17}_{-9}$ ms for the $(3/2^-)$ ground state and $E_\alpha = 7612(8)$ keV, $T_{1/2} = 24^{+6}_{-4}$ ms for the $(13/2^+)$ isomeric state [31]. The α_3 event is an escape decay and its full energy can be recovered by summing up the two energies deposited in the PSSD (5461 keV) and the side detector (1570 keV). The obtained energy of 7031 keV and decay time of 0.316 s are compatible with the α-decay properties of ^{199}Rn, $E_\alpha = 6989(6)$ keV, $T_{1/2} = 0.59(3)$ s for the $(3/2^-)$ state and $E_\alpha = 7060(6)$ keV, $T_{1/2} = 0.31(2)$ s for the $(13/2^+)$ state [32]. In particular, the α_4 decay can only be associated with the $(3/2^-)$ ground state of ^{195}Po for which $E_\alpha = 6606(5)$ keV and $T_{1/2} = 4.64(9)$ s [32]. Therefore, we conclude that the parent nucleus is the new isotope ^{207}Th and the observed decay chain is of the type ER-α_1(^{207}Th)-α_2(^{203g}Ra)-α_3(^{199g}Rn)-α_4(^{195g}Po). Based on this chain, the α-particle energy and half-life of ^{207}Th were determined to be 8167(21) keV and $9.7^{+46.6}_{-4.4}$ ms. The half-life was extracted using the exact method described in Ref. [30]. The production cross section of ^{207}Th was estimated to be 4^{+9}_{-3} pb.

In addition, three α-decay chains of the known isotope ^{208}Th were also observed in this experiment (see Fig. 1). On the basis of these events, an α-particle energy of 8053(18) keV and a half-life of $4.4^{+6.0}_{-1.6}$ ms were deduced for ^{208}Th, which agree with the previously reported data of $E_\alpha = 8044(30)$ keV and $T_{1/2} = 1.7^{+1.7}_{-0.6}$ ms [33]. As illustrated in Fig. 2(a), the newly measured α-decay energies of ^{207}Th [8328(21) keV] and ^{208}Th [8211(18) keV] fit well into the systematics of the Q_α values of ground-state to ground-state transitions, which indicate that the observed α decays originate from ground states. The experimental Q_α values for nuclei with $Z > 82$ and $N \leqslant 126$ behave quite regularly as shown in Fig. 2(a). From the systematics, a global trend that Q_α in each isotopic chain increases as a function of decreasing neutron number can be deduced. A noticeable feature of the global trend is that, starting from $N = 125$ downward, the Q_α values along isotopic chains actually follow a stepwise rather than smooth upward trend, namely, an odd-N isotope has a smaller Q_α value than the average of its two even-N neighbors. A similar situation is also observed in isotonic chains [see Fig. 2(d)], but here, the Q_α value of an odd-Z isotone is larger than the average of its two adjacent even-Z isotones. It is definite that there exists OES in α-decay energies for nuclei with $Z > 82$ and $N < 126$ along both isotopic and isotonic chains.

To study this phenomenon quantitatively, the OES of α-decay energies is defined as the commonly used three-point indicator [8–10],

$$\Delta Q(A) = \tfrac{1}{2}[2Q_\alpha(A) - Q_\alpha(A-1) - Q_\alpha(A+1)], \quad (1)$$

where $Q_\alpha(A)$ is the α-decay energy of a nucleus with mass number A. When the number of protons or neutrons is fixed, Eq. (1) gives the OES along an isotopic chain (neutron OES $\Delta Q_{\rm n}$) or isotonic chain (proton OES $\Delta Q_{\rm p}$). Values of $\Delta Q_{\rm n}$ and $\Delta Q_{\rm p}$ extracted from the experimental Q_α values are shown in Figs. 2(b) and 2(e), respectively. It is clear that with a few

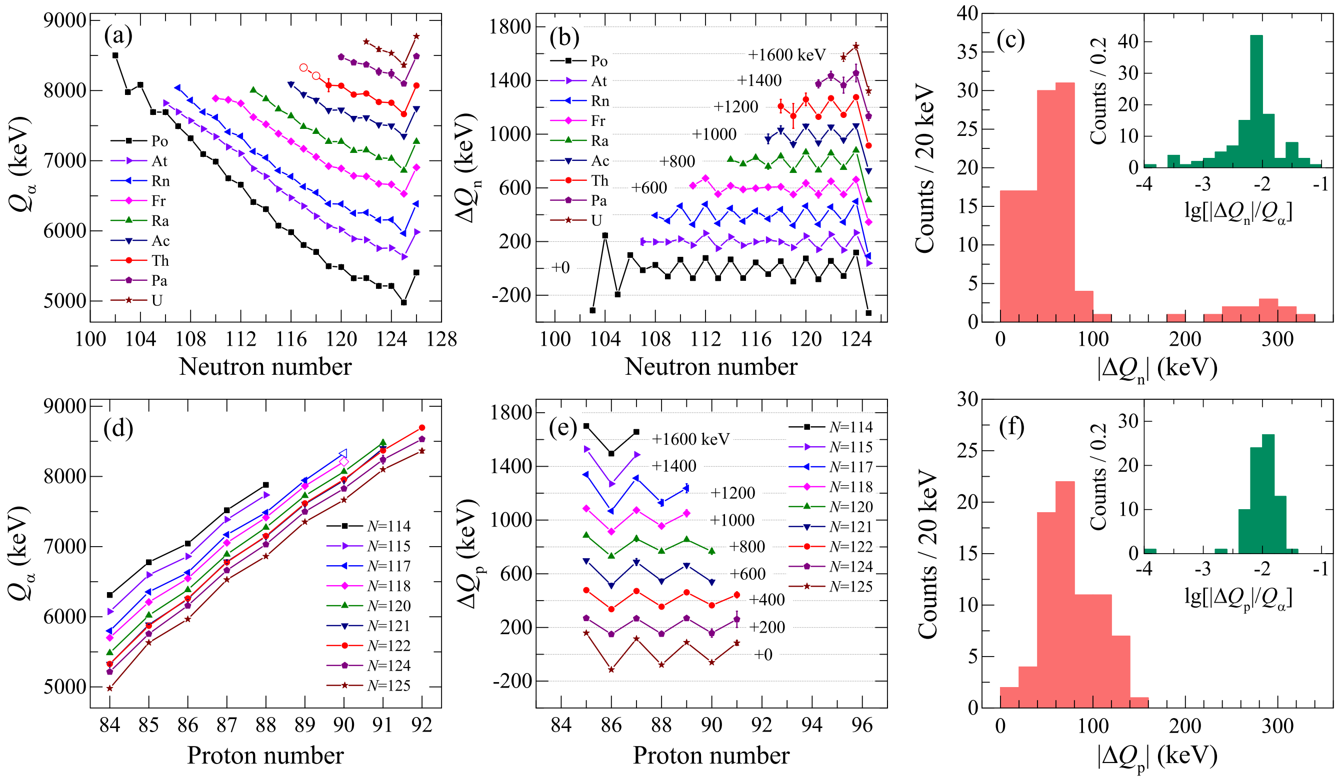

FIG. 2. The OES in α-decay energies. (a) The Q_α values of ground-state to ground-state transitions for nuclei with $Z > 82$ and $N \leqslant 126$ along isotopic chains. Open symbols refer to the values of ^{207,208}Th measured in this Letter. Solid symbols are experimental data taken from relevant literature [19,20,22,34–43] except for the value of ^{209}Th, which is a predicted value from Ref. [44]. (b) The values of ΔQ_n evaluated using Eq. (1) are plotted against neutron number. The curves are shifted by certain values avoiding overlapping. (c) The distribution of $|\Delta Q_n|$. The inset displays the ratios of $|\Delta Q_n|$ to Q_α on the logarithmic scale. (d)–(f) The same as (a)–(c) but for data along partial isotonic chains.

exceptions both ΔQ_n and ΔQ_p exhibit regular patterns, i.e., alternating positive and negative values occur in every isotopic and isotonic chains. Regardless the error bars, the absolute values of ΔQ_n are mainly in the range of 0–120 keV with the mean value of 72 keV [see Fig. 2(c)]. The ratios of $|\Delta Q_n|$ to Q_α are mostly in the range of 1.0×10^{-3}–4.0×10^{-2} with the mean value of 1.06×10^{-2}. Compared to the trend of ΔQ_n, the behavior of ΔQ_p shown in Fig. 2(e) exhibits a better regularity. The absolute values of ΔQ_p are ranged from 20 to 160 keV with the mean value of 76 keV [see Fig. 2(f)]. The ratios of $|\Delta Q_p|$ to Q_α have a range of 4.0×10^{-3}–2.5×10^{-2} with the mean value of 1.1×10^{-2}. On the whole, the typical amplitude of the OES in α-decay energies for nuclei with $Z > 82$ and $N < 126$ is about 70 keV, which accounts for ~1% of the α-decay energies. However, according to the classical Bethe-Weizsäcker mass formula [45], the calculated OES of α-decay energies for nuclei in this region is only 3–12 keV in magnitude, which is much smaller than the experimental values. In addition, the calculated proton OES even presents an opposite trend to the experimental one. Certainly, the OES in α-decay energies could not be reproduced by the Bethe-Weizsäcker formula.

To illustrate the significance of the OES in α-decay energies, we compare this effect to the well-known OES in nuclear binding energies (B) for the same set of nuclei. Applying Eq. (1) to the experimental binding energies [36], i.e., replacing the Q_α in Eq. (1) with B, we readily obtain the neutron OES ΔB_n and proton OES ΔB_p of binding energies. The absolute values of ΔB_n are close to that of ΔB_p, and they lie between 0.5 and 1.5 MeV. Such values only account for 0.04%–0.09% of the binding energies. It is evident that the values of $|\Delta Q_n|/Q_\alpha$ and $|\Delta Q_p|/Q_\alpha$ are almost an order of magnitude larger than those of $|\Delta B_n|/B$ and $|\Delta B_p|/B$, namely, the effect of OES on α-decay energies is even more significant than that on binding energies. Therefore, contrary to popular belief, we believe that the OES in α-decay energies is a regular and distinct but not negligible effect.

In order to probe the underlying mechanism of the OES in α-decay energies, we performed both RHFB and LSSM calculations from which the theoretical binding energies, α-decay energies and OES were obtained. The RHFB calculation was performed using the effective interaction PKA1 [46] for the mean field and the Gogny interaction for the pairing field with adjusted pairing strength based on D1S force [47]. The LSSM calculation was carried out using a Hamiltonian constructed from the modified Kuo-Herling interaction and monopole based universal interaction [22,48–50]. As examples, the OES of Th isotopes and $N = 117$ isotones from the RHFB calculation, and the OES of Po isotopes and $N = 125$ isotones from the LSSM calculation are presented in Figs. 3 and 4 together with experimental data, respectively. In the RHFB calculation, the ground states of Th isotopes with odd neutron number are obtained by blocking neutron $2f_{5/2}$ orbital (i.e., the valence neutron occupies the $2f_{5/2}$ orbital) if $N < 118$,

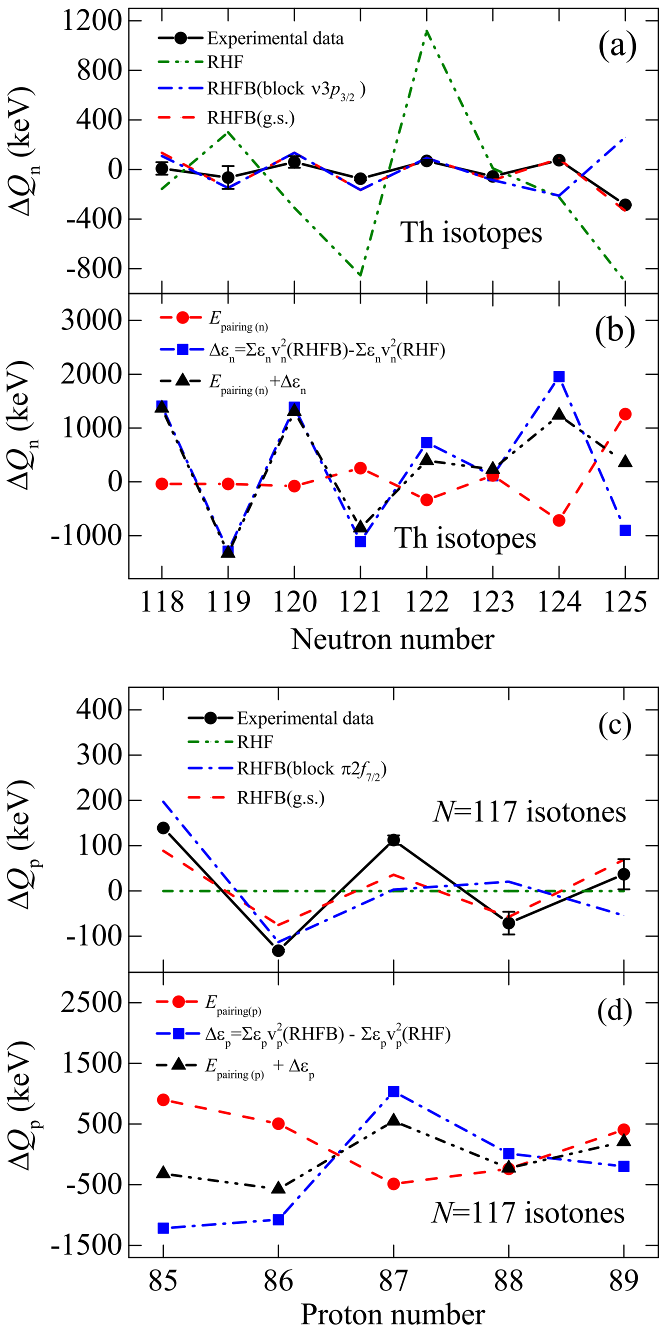

FIG. 3. Theoretical results obtained from the RHFB calculations. (a) Comparison of experimental and theoretical OES values for Th isotopes. The red dashed and blue dashed-dot lines show the RHFB results with blocking of the orbital that gives the ground state and the specific orbital, respectively. The RHF calculation with blocking of the ground-state orbital is presented by the olive dashed-dot-dot lines. (b) The contribution to OES from the pairing energy E_{pairing} (red circle), the change in total single-particle energy caused by pairing scattering $\Delta\varepsilon$ (blue square), and the sum (black triangle) of E_{pairing} and $\Delta\varepsilon$ for Th isotopes. (c) and (d) The same as (a) and (b) but for $N = 117$ isotones.

the $3p_{3/2}$ orbital if $118 < N < 124$ and the $3p_{1/2}$ orbital if $N > 124$. For $N = 117$ isotones, the ground states are obtained by blocking neutron $3p_{3/2}$ orbital and proton $1h_{9/2}$ orbital for the odd proton number. Their daughter nuclei have

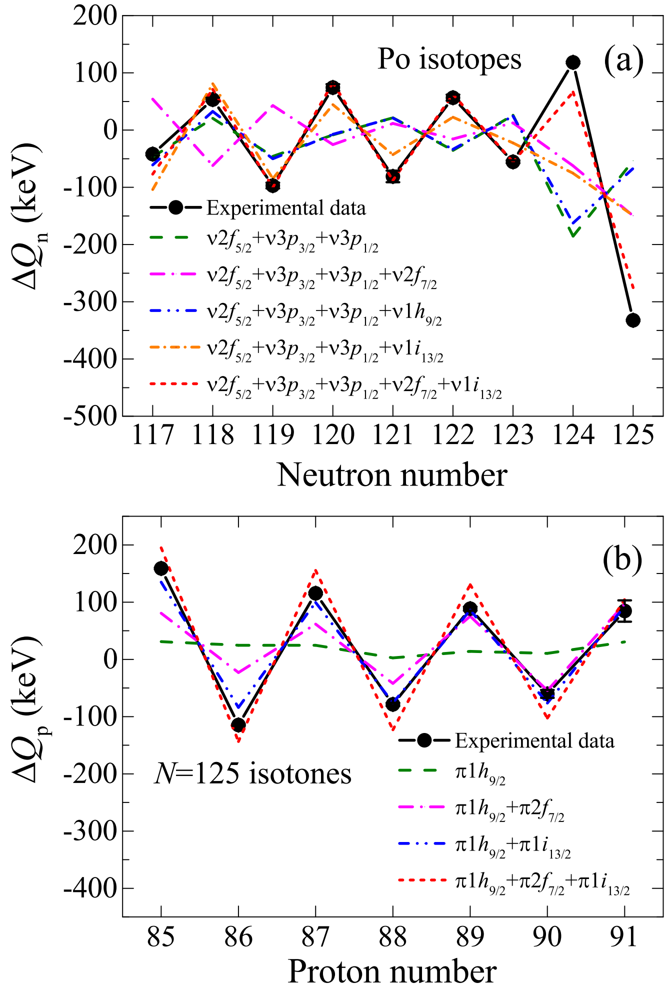

FIG. 4. Theoretical results obtained from the LSSM calculations. (a) Comparison of experimental and theoretical OES values for Po isotopes. (b) Comparison of experimental and theoretical OES values for $N = 125$ isotones. Lines in (a) and (b) represent the results obtained by constraining nucleons in different orbitals.

the same way of blocking. As shown in Figs. 3(a) and 3(c), the calculations (red dashed lines) well reproduce the OES of both Th isotopes and $N = 117$ isotones. For comparison, Figs. 3(a) and 3(c) also present the results calculated without pairing correlations, namely, calculated by the RHF model (olive dashed-dot-dot lines) with the same blocking, and the results of RHFB calculation with blocking of neutron $3p_{3/2}$ orbital for Th isotopes and the proton $2f_{7/2}$ orbital for $N = 117$ isotones (blue dashed-dot lines). One can see that without the inclusion of pairing correlations, the theoretical staggering becomes irregular for Th isotopes, and even completely disappears for $N = 117$ isotones. On the other hand, even if pairing correlations are included but the blocking of orbitals is changed, the calculations could not reproduce the experimental OES well. Therefore, we conclude that both the pairing correlations and blocking of a specific orbital for unpaired nucleon are essential for the occurrence of OES in α-decay energies.

Furthermore, pairing correlations show their effects in two ways. One is the pairing energy released in the formation of a Cooper pair, and the other one is the pairing scattering, i.e., nucleons are scattered to orbitals above Fermi levels. To explore these two effects on the occurrence of OES, we further investigate the contributions to OES from the neutron (proton) pairing energy E_{pairing}, the changes of total neutron (proton) single-particle energy $\Delta\varepsilon$ caused by pairing scattering, namely, the total neutron (proton) single-particle energy difference between RHFB and RHF calculations with the same blocking for the odd nucleon, and the sums of E_{pairing} and $\Delta\varepsilon$ for Th isotopes and $N = 117$ isotones. The results are presented in Figs. 3(b) and 3(d). It can be seen that for Th isotopes the pairing scattering is the main cause of the OES, whereas for $N = 117$ isotones both pairing energy and pairing scattering are important for the occurrence of OES. Therefore, the pairing correlations influence the OES not only just through the contribution of pairing energy to the binding energy, but also by scattering nucleons to orbitals away from Fermi levels. In the LSSM, the calculations are carried out in model space with the same proton and neutron single-particle orbitals, including $1h_{9/2}$, $2f_{7/2}$, $2f_{5/2}$, $3p_{3/2}$, $3p_{1/2}$, and $1i_{13/2}$. We use the Po isotopes and $N = 125$ isotones as examples to demonstrate the calculated results. For Po isotopes, protons are constrained in $1h_{9/2}$, $2f_{7/2}$, and $1i_{13/2}$ orbitals, and neutron holes are constrained in five sets of orbitals. The results are compared with experimental data in Fig. 4(a), it is clear that neutron $1i_{13/2}$ and $2f_{7/2}$ orbitals are crucial to reproduce the observed OES of Po isotopes, although the $2f_{5/2}$, $3p_{3/2}$, and $3p_{1/2}$ orbitals are closer to the $N = 126$ shell closure and dominate hole configurations. Similar results are also obtained for the $N = 125$ isotones where neutron holes are constrained on $2f_{5/2}$, $3p_{3/2}$, and $3p_{1/2}$ orbitals and protons are constrained in four sets of orbitals. As shown in Fig. 4(b), the $1i_{13/2}$ orbital still plays a key role to reproduce the observed OES of the $N = 125$ isotones, even though protons are expected to occupy $1h_{9/2}$ orbital first [22]. Obviously, the LSSM calculations can exactly reproduce the experimental OES on the condition that a single nucleon occupies the specific orbital and concurrently more nucleons should be involved in the configuration mixing. It is worth mentioning that both the configuration mixing in LSSM and pairing scattering in RHFB are beyond mean-field effects, and the configuration mixing in LSSM can be considered as a natural consequence of the pairing scattering in RHFB.

Based on the above discussions, we find that the observed OES in α-decay energies reveals a novel mechanism induced by pairing scattering or complex configuration mixing, which is not included in the classical Bethe-Weizsäcker formula. Both the specific orbitals near the Fermi surface and the orbitals away from the Fermi surface, especially the unnatural parity orbital from spin-orbit splitting (herein $1i_{13/2}$), are of particular importance to describe the OES in α-decay energies.

Overall, a new α-emitting thorium isotope ^{207}Th has been identified with an α-particle energy of 8167(21) keV and a half-life of $9.7^{+46.6}_{-4.4}$ ms. The previously reported α-decay properties of the neighbor isotope ^{208}Th have been confirmed by observing three α-decay chains. Combining the newly measured α-decay energies with existing data, we obtained an updated systematics of α-decay energies for nuclei with $Z > 82$ and $N < 126$. From the systematics, it was strikingly found that there is a regular and distinct OES in α-decay energies along both isotopic and isotonic chains, and the magnitude of the OES is typically in the range of tens to more than 100 keV, which is about an order of magnitude larger than the values deduced from the classical Bethe-Weizsäcker formula. This finding clearly indicated that the OES in α-decay energies is a regular and distinct effect rather than a negligible one as commonly supposed. The OES was well reproduced by both the RHFB and LSSM calculations. Accordingly, we proposed that the OES in α-decay energies originates from both pairing correlations and blocking effect of unpaired nucleons. The pairing correlations bring about the OES not only through the contribution of pairing energy to binding energy but also by configuration mixing resulting from scattering of nucleons to orbitals away from Fermi levels.

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