Reinvestigation of ^{222}U in high-precision digital α-decay spectroscopy: Solution to the reduced decay-width anomaly

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The α decay of ^{222}U was reinvestigated using a general least-square superpulse fitting algorithm dedicated to resolving pileup signals in the decay of very short-lived nuclei. The α-particle energy of ^{222}U was revised to be 9246(8) keV, and the precision was improved significantly compared with previous result. Using the present α energy, the anomaly in the systematics of the α-decay reduced width δ^2 observed at ^{222}U in the N_pN_n scheme is solved, all the δ^2 values converge into a smooth and narrow band with N_pN_n up to $\approx$90 in the northeast of ^{208}Pb.

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I. INTRODUCTION

Shell evolution far from the stability line is one of the frontiers in temporary nuclear physics research [1,2]. Being the heaviest nuclear magic number, the robustness of the $N = 126$ shell closure has been a subject of intense investigation and has been probed beyond the proton dripline up to neptunium ($Z = 93$) [3–5]. Due to very low production yields, experimental data in this region are very scarce and were obtained mainly in α-decay spectroscopy. Information on nuclear structure can be obtained from the α preformation probability [6] or the equivalent reduced α-decay width δ^2 [7], which microscopically quantify the stability against α decay. In our recent work, the systematics of decay widths around $N = 126$ above Pb ($Z = 82$) was investigated and found remarkably simplified in the N_pN_n scheme, especially for those in the northeast of ^{208}Pb. The correlation between δ^2 and N_pN_n is extraordinarily compact, with ^{222}U standing out as an only exception (see Fig. 3(b) in Ref. [8]).

Nuclei in this region are very short lived, especially for α emitters with $Z \geqslant 84$ and $N = 128$–130 due to the effect of the $N = 126$ shell closure, with half-lives in the range of nanoseconds to microseconds. In their implantation decay correlation measurements with the conventional analog electronics, signal pileup becomes an issue as met in the first discovery experiment of ^{222}U [9], causing the energy and decay time information to be distorted or even missed.

The digital pulse processing (DPP) technique was introduced by recording the waveforms of the signals [10], and first successfully applied to resolve pileup events in the decay of short-lived proton emitters ^{144,145}Tm [11,12] and the fast α decays above the self-conjugate doubly magic ^{100}Sn [13]. This technique introduces much flexibility as the digitized waveforms can be analyzed off line by using various pulse shape analysis algorithms and has become indispensable in the study of the most exotic p-rich isotopes such as ^{149}Lu [14], ^{185}Bi [15], and the heaviest self-conjugate isotopes ^{104}Te and ^{108}Xe [16].

In our recent studies in the heaviest proton dripline region around $N = 126$ carried out at the gas-filled recoil separator SHANS [17], fully digital focal plane detection setups using double-sided silicon strip detector (DSSD) [18–21] or position-sensitive silicon strip detectors (PSSDs) [3,5,22,23] as implantation detector were exploited. Different algorithms were developed for the pulse shape analysis of DSSD [18,24] and PSSD [25] signals, respectively.

Following the superpulse scheme proposed for the analysis of DSSD waveforms in Ref. [26], our collaboration developed two algorithms [24,27] to decompose pileup events by fitting the superpulse to the experimental traces. In Ref. [24], with an exponential function baseline correction and plateau-region fitting, the spectroscopic information of pileup signals with time separation down to 80 ns and amplitude of

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overlapping pulse down to 70 keV, can be readily extracted. In the implementation of this algorithm, the fitting is done plateau by plateau, i.e., signal by signal, skipping the leading edges, leading to large uncertainties in the characterization of signals associated with short-length plateaus. Therefore it was refined in two aspects [27]. First, a combination of a linear function and an exponential function was introduced to account for the baseline more precisely. Second, the amplitude and arrival time of each individual signal are obtained simultaneously in global weighted least-square fitting, including the leading edges. The weight at each bin is inversely proportional to the uncertainty of the superpulse, with the largest uncertainties and therefore the smallest weights in the leading edges. The initial value of the arrival time of each individual signal is taken as the peak time of the symmetrical triangular filter. This new development [27] shows overall performance comparable with Ref. [24], the energy resolution for overlapping signals with time difference shorter than 200 ns slightly improved. Detailed descriptions of the algorithm and performance validation are given in Ref. [27].

In our recent experiments at SHANS, very short-lived nuclei with $N = 128$–130, including ^{222}U, were produced either as evaporation residues (ERs) or as decay products of ERs [18–23]. In this paper, we report on the new results for the α decay of ^{222}U obtained using the general least-square superpulse fitting algorithm [27].

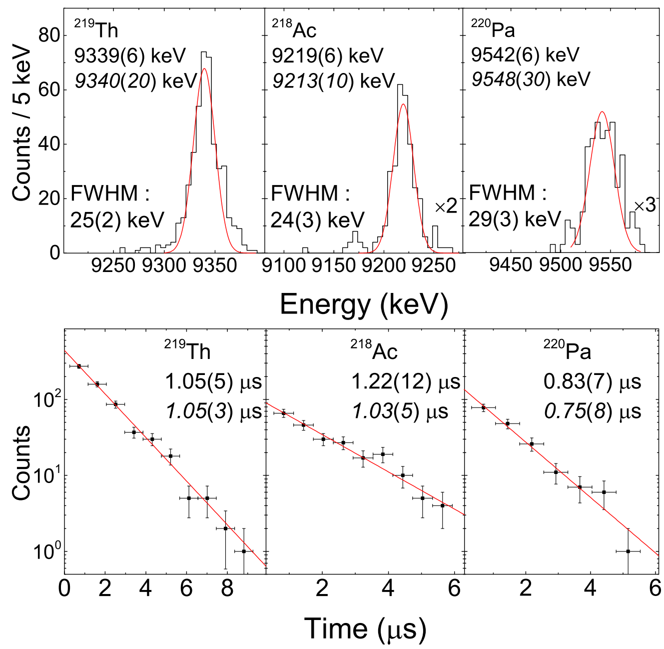

FIG. 1. The α energies, half-lives of ^{219}Th, ^{218}Ac, and ^{220}Pa obtained from the present digital signal waveform analysis. The α energy and half-life values in italics below our results are from Refs. [32], [33], and [23], respectively.

II. EXPERIMENT

The experiments were performed at the Heavy Ion Research Facility in Lanzhou (HIRFL). ^{222}U was produced in the fusion evaporation reaction induced by a 188 MeV ^{40}Ar beam on a 467μg/cm^2 enriched ^{186}W target with 80 μg/cm^2 carbon backing via the ^{186}W(^{40}Ar, 4n) reaction channel. ERs were separated from the primary beam by the He-filled recoil separator SHANS [17] and implanted into a 128 × 48 300-μm DSSD. The DSSD had 48 horizontal and 128 vertical strips of 1 mm width, providing a total of 6144 effective pixels. The identification of the decay chains was achieved by using standard position-and-time correlation method [28]. Signals from the preamplifiers of the DSSD were digitized by the 14 bit, 100 MHz CAEN V1724 fADC modules [29]. The preamplifier signals [30] with rise time and decay time of about 40–60 ns and 300 μs, respectively, were sampled at a frequency of 50 MHz. Each recorded trace was 15 μs long. More details about the experimental setup can be found in Ref. [18]. The construction of the superpulse was carried out in the same way as in Ref. [24] with internal α decays of long-lived evaporation residues produced in the ^{40}Ar + ^{175}Lu reactions. A beam with an intensity of 500 particle nA was delivered to the target over a period of 9 h.

Energy calibration was performed in the energy range of 6.0–9.6 MeV for the new algorithm, using single signal α waveforms registered with the ^{175}Lu target and single and double signal waveforms (^{215}Ra and ^{219}Th) recorded with the ^{186}W target. A systematic energy uncertainty of 5.5 keV was deduced from the calibration following the method in Ref. [31]. The energy uncertainties were calculated as the quadrature of the statistical and systematic errors.

III. RESULTS

Three short-lived $N = 129$ isotones ^{219}Th, ^{218}Ac, and ^{220}Pa with half-lives around 1μs are chosen as benchmarks and their α spectra and half-lives deduced with the new algorithm are displayed in Fig. 1. For very fast decays with time separation in the range of 80–500 ns between the ER and subsequent α-decay signals, energy resolutions (FWHM) of around 29 keV are obtained, compared to 32 keV achieved in Ref. [24]. In the GSI work [4], where the α energy of ^{222}U was first measured with digital spectroscopy, the best energy resolutions of multiple α events stored in a single trace with time differences down to 1 μs and 0.17 μs were ≈110 keV and ≈180 keV, respectively.

Traces followed by α decay of ^{214}Ra [$E_\alpha = 7137(3)$ keV, $T_{1/2} = 2.46(3)$ s] [34], the third member of the α-decay chain originating from ^{222}U, with time difference shorter than ten times the half-life of ^{214}Ra, were checked event by event. Nine traces with three signals labeled as ERs and one with two decay signals preceded by an ER event (chain No. 3 in Table I) were found and attributed to the implantation of ^{222}U followed by α decays of ^{222}U and ^{218}Th. The corresponding decay chains are listed in Table I. The trace corresponding to event 5 is plotted in Fig. 2 as an example.

The α energies of ^{222}U in five of the decay chains are very similar and their average was taken as full energy, 9246(8) keV. It is at variance with the value of 9310(50) keV obtained in the GSI experiment using DSSD as implantation detector, where 81 triple (ER-α_1-α_2) traces were attributed to ^{222}U and a quite conservative energy uncertainty was adopted. The half-life of ^{222}U was determined to be $4.0^{+1.9}_{-1.0}$ μs following the method in Ref. [35], consistent with the GSI result [4]. Based

TABLE I. The decay chains assigned to ^{222}U in the present work.

	ERs	^{222}U		^{218}Th		^{214}Ra	
Chain No.	E (MeV)	$E_{\alpha 1}$ (keV)	$\Delta t_{\alpha 1}$ (μs)	$E_{\alpha 2}$ (keV)	$\Delta t_{\alpha 2}$ (ns)	$E_{\alpha 3}$ (keV)	$\Delta t_{\alpha 3}$ (s)
1	15.00	634[a]	8.56	2238[a]	402	7129	1.02
2	13.64	1569[a]	0.57	9711	104	7123	3.57
3	14.91	9229	25.45	9706	495	7134	7.68
4	15.48	9265	11.52	9697	272	7111	0.75
5	14.31	9245	1.58	9676	343	7128	2.11
6	11.66	9248	1.95	9624[d]	327	7116	1.04
		(5314+3934)[b]		(9480+144)[b]			
7	11.65	11780[c]	2.99	–	37	7140	4.53
8	12.30	10100[c]	0.65	–	63	7131	1.44
9	13.95	9244	2.14	339[a]	191	7151	0.77
		(7745+1499)[b]		(284+55)[b]			
10	13.07	10217[c]	2.74	–	<20	7136	4.71
This work			$T_{1/2}$/μs		$T_{1/2}$/ns		$T_{1/2}$/s
		9246(8)	$4.0^{+1.9}_{-1.0}$	9697(10)	155^{+72}_{-37}	7130(6)	$1.91^{+0.89}_{-0.46}$

[a]Partial energy escaped.
[b]Reconstructed from two adjacent strips.
[c]The two signals were inseparable due to the very short time difference.
[d]Partial energy lost because of overshoot.

on the present results, the reduced decay width of ^{222}U was deduced to be 160^{+76}_{-40} keV.

For ^{218}Th, an energy of 9697(10) keV was obtained from the average for four decay chains (Nos. 2–5), decay chain 8 was excluded as the time difference between the two overlapping α signals is too short for our algorithms [24,27]. It is in disagreement with the evaluated value [33], but consistent with one of the earliest results [36] and the latest obtained in digital α spectroscopy [4,37], see Table II.

In decay chain 10, only an upper limit can be given for the decay time of ^{218}Th, so a statistical approach to lifetime measurement with restricted observation times was applied [38]. This case belongs to the emerging scenario, from which a half-life with mean value 155(49) ns can be estimated by frequentist inference with $T = 20$ ns, $\bar{t} = 248$ ns, $n = 1$, $m = 9$. The half-life of ^{218}Th was thus extracted to be 155^{+72}_{-37} ns, consistent with the evaluated value [33] (see Tables I and II). Based on our results, the reduced decay width of ^{218}Th is deduced to be 89^{+42}_{-21} keV, consistent with the value ($\delta^2 =$ 132(5) keV) obtained using the evaluated E_α and $T_{1/2}$ values [33].

In order to get reliable structure information, it is often necessary to combine data from different experiments, as done for ^{216}U in Ref. [8]. Similarly, using the energies measured in the present work and the combined half-lives measured here and in previous works, the α-decay reduced widths of ^{222}U and ^{218}Th are deduced to be 139(20) keV and 111(4) keV, respectively, as presented in Table II.

The N_pN_n scheme provides a simple, but powerful, guide to understanding and predicting the systematic behavior of nuclear properties. It has been demonstrated with different physical variables, such as the excitation energy $E(2^+)$, the ratio of the excitation energies $E(4^+)/E(2^+)$ and reduced transition rate $B(E2)$ of first 2^+ states in even-even nuclei, that extremely complex systematics seen in normal plots against neutron number N or proton number Z are significantly simplified and the data coalesce into smooth and narrow bands in N_pN_n plots [43,44]. In Fig. 3, the reduced decay widths of

TABLE II. The g.s.-to-g.s. α-decay energies and half-lives of ^{222}U and ^{218}Th measured in this work. The reduced α-decay widths δ^2, in column 4, are calculated by Rasmussen formalism [7]. The data for ^{222}U and ^{218}Th are compared with literature values.

	This work			Literature data		
Isotope	E_α (keV)	$T_{1/2}$(μs)	δ^2 (keV)	E_α (keV)	$T_{1/2}$(μs)	Ref.
^{222}U	9246(8)	4.62(66)[a]	139(20)	9310(50)	4.7(7)	[4,39]
^{218}Th	9697(10)	0.122(5)[a]	114(5)	9668(10)	0.122(5)	[33]
				9680(20)	0.122(8)	[36]
				9665(10)	0.096(7)	[40,41]
				–	0.125(5)	[42]
				9670(80)	0.16(4)	[4]
				9720(70)	$0.169^{+0.073}_{-0.040}$	[37]

[a]Combined half-life from the present and previous work(s) using weighted average.

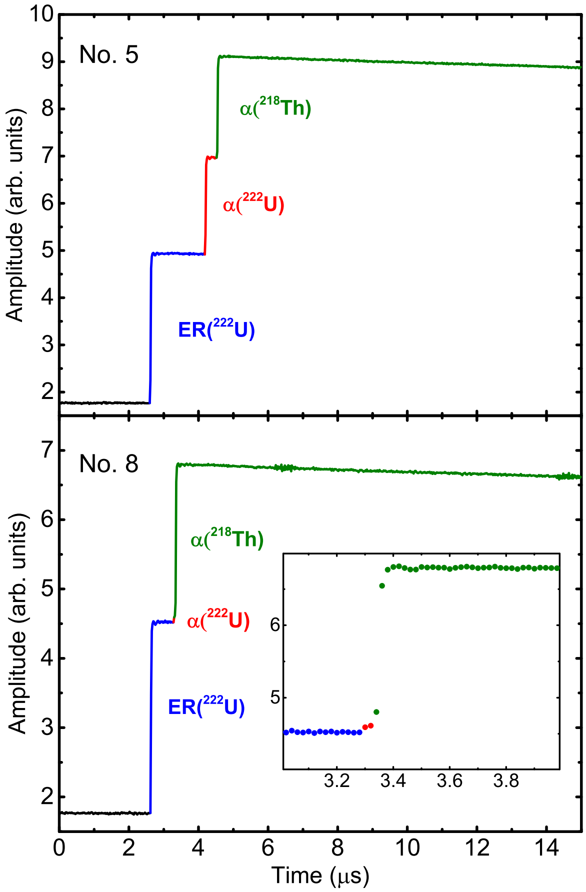

FIG. 2. The waveforms for chains 5 and 8 in which ^{222}U implant and subsequent α decays of ^{222}U and ^{218}Th were registered. The inset shows the zooming in of the $\alpha(^{222}$U) region, where only the sum energy can be extracted for too close pileup signals.

even-even nuclei in the northeast of ^{208}Pb are displayed with the revised results for ^{222}U and ^{218}Th, while the values for others are the same as in Fig. 3 of Ref. [8]. Now ^{222}U follows the overall systematics in this region rather well, therefore the anomaly at ^{222}U is solved.

The δ^2 systematics is strikingly simple while the explanation remains to be a challenge for theory. The reduced decay width is influenced by many factors, which are often intertwined inseparably, such as pairing force and n-p interaction acting between the neutrons and the protons that constitute the α particle and structure change between mother and daughter nuclei. To uncover the underlying mechanism it needs breakthrough, which has been waited for a quarter century since the seminal review on cluster radioactivity theory [45].

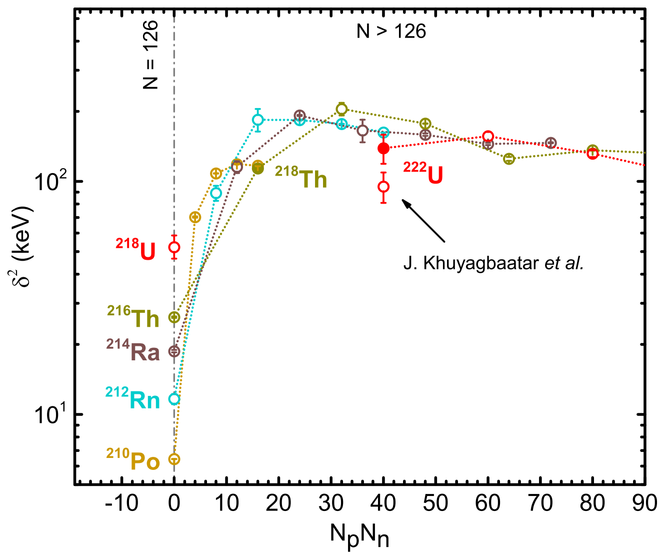

FIG. 3. Systematics of reduced decay widths for g.s.-to-g.s. α decays of even-even $84 \leqslant Z \leqslant 92$ and $N \geqslant 126$ isotopes as a function of N_pN_n. The values for ^{222}U and ^{218}Th are deduced using the α energies from this work and the combined half-lives as shown in Table II and are displayed with filled circles. The result of ^{222}U from the work of Khuyabaatar *et al.* [4] is shown as an open circle. The errors of reduced decay widths are determined with half-life uncertainties only.

IV. CONCLUSION

In summary, the α decay of ^{222}U involving the pileup of $ER(^{222}\text{U})$-$\alpha_1(^{222}\text{U})$-$\alpha_2(^{218}\text{Th})$ was reinvestigated using the refined general least-square superpulse fitting algorithm. The α energy and reduced decay width of ^{222}U were revised to be 9246(8) keV and 143^{+18}_{-12} keV, respectively. The anomaly in δ^2 observed at ^{222}U in the N_pN_n scheme is solved, all the δ^2 values form an extraordinarily compact trajectory to $N_pN_n \approx 90$ in the northeast of ^{208}Pb.

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