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Fine structure in the α decay of the 8^+ isomer in ^{216,218}U

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The extremely neutron-deficient even-even uranium isotopes ^{216,218}U were produced in the complete-fusion reactions induced by impinging ^{40}Ar and ^{40}Ca ions on ^{180,182,184}W targets. Fusion evaporation residues were separated in flight by the gas-filled recoil separator SHANS (Spectrometer for Heavy Atoms and Nuclear Structure) and subsequently identified using the recoil-α-correlation method. The improved ground-state to ground-state α-decay properties of ^{216,218}U were reported in [Z. Y. Zhang *et al.*, Phys. Rev. Lett. **126**, 152502 (2021)]. In this paper, we report on new α-decay activities with $E_\alpha = 10\,163(27)$ keV for ^{216}U and $E_\alpha = 10\,073(16)$ keV for ^{218}U, which decay from the 8^+ isomeric states of ^{216,218}U into the 2^+ states of their daughter nuclei ^{212,214}Th, respectively. The new results extend the systematics of the α-decay fine structure for the $N = 124$ and 126 even-even isotones.

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I. INTRODUCTION

Alpha-decay spectroscopy is a powerful tool to identify new isotopes and to investigate the nuclear structure and masses of ground and excited states in neutron-deficient heavy-mass regions [1–3]. For nuclei with neutrons below $N = 126$ and protons above $Z = 82$, the α-decay studies are challenging due to their tiny production cross sections and short half-lives. In the case of the extremely neutron-deficient even-even uranium isotopes ^{216}U [4–6] and ^{218}U [7–9], the ground state (g.s.) α-decay properties were determined based on only a few α-decay chains, resulting in large uncertainties on decay energies and half-lives. In addition, α-decaying isomeric states with spin and parity of $J^\pi = 8^+$ were observed in both ^{216}U and ^{218}U, to which the presumed configuration of $\pi h_{9/2} f_{7/2}$ was assigned [5,9]. Prior to our study, only the α decays from the 8^+ isomeric states directly to the corresponding 0^+ ground states of daughter nuclei ^{212,214}Th were observed experimentally [5,9]. However, in the lighter even-even $N = 126$ isotones ^{214}Ra and ^{216}Th, fine structure in the α decay from the 8^+ isomer, i.e., $8^+ \to 0^+$, $8^+ \to 2^+$, and $8^+ \to 8^+$ α decays, was observed [10,11]. The same α-decay pattern is expected in their isotones; therefore, it is interesting to search for the fine structure in α decays of ^{216m,218m}U.

In this work, the α-decay properties of ^{216,218}U were measured with significantly improved statistics. The g.s. to g.s.

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α-decay data were reported in Ref. [12]. Here, we report on new α radioactivities from ^{216m}U and ^{218m}U to the corresponding 2^+ states of their daughter nuclei ^{212,214}Th.

II. EXPERIMENTAL DETAILS

Three separate measurements employing the ^{40}Ar + ^{182}W, ^{40}Ca + ^{184}W, and ^{40}Ar + ^{180}W reactions were performed at the gas-filled recoil separator SHANS (Spectrometer for Heavy Atoms and Nuclear Structure) [13]. The ^{40}Ar and ^{40}Ca beams with a typical intensity of 200–700 pnA were delivered by the Sector-Focusing Cyclotron of the Heavy Ion Research Facility in Lanzhou (HIRFL), China. The isotope ^{218}U was produced in the complete fusion reactions ^{182}W(^{40}Ar, $4n$)^{218}U and ^{184}W(^{40}Ca, $\alpha 2n$)^{218}U. Beam energies were 190 MeV for ^{40}Ar and 206 MeV for ^{40}Ca, respectively. For ^{216}U, the ^{180}W(^{40}Ar, $4n$)^{216}U reaction was used, with the beam energy of 191 MeV. The targets of ^{180,182,184}W (enrichment of 91.4% for ^{180}W, 91.4% for ^{182}W, and 96.3% for ^{184}W) with thicknesses of 260–500 μg/cm^2 were made by sputtering the material onto an 80 μg/cm^2 thick carbon foil and then covered by a 10 μg/cm^2 thick carbon layer.

The gas-filled recoil separator SHANS was used for the separation of recoiled evaporation residues (ERs) from the primary beam particles and other unwanted reaction products. The separator was filled with helium gas at a pressure of 0.6 mbar and the magnets were set to guide the ERs to the center of the focal plane. ERs surviving during the flight were implanted into three 300-μm-thick position-sensitive strip detectors (PSSDs) installed side by side at the focal plane of the separator. Each PSSD with an active area of 50 × 50 mm^2 was divided into 16 vertical strips on the front surface. To detect the α particles that escaped from the PSSDs, eight non-position-sensitive side silicon detectors (SSDs) with 50 × 50 mm^2 size were mounted perpendicular to the surface of the PSSDs in an open box geometry. The efficiency of the detector array for the detection of the full-energy α particles was measured to be 72%. In order to distinguish the α-decay events from the implantation ones, two multiwire proportional counters were mounted 15 and 25 cm upstream from the PSSDs. Behind the PSSDs, three punch-through detectors were placed to provide veto signals for energetic light particles passing through the PSSDs. The transport efficiency of SHANS was estimated to be 14% by using the reaction ^{40}Ar + ^{175}Lu [13]. Signals from the preamplifiers of the detectors were recorded employing a digital data acquisition system, which consists of 16 wave-form digitizers V1724 from CAEN S.p.A [14]. More details of the system are given in Refs. [15–17].

The energy calibration of PSSDs and SSDs was performed using a three-peak (^{244}Cm, ^{241}Am, and ^{239}Pu) α source and the known α lines from ^{205,206}Rn, ^{208,209}Fr, and ^{210,211}Ac, which were produced in the test reactions of ^{40}Ar + ^{175}Lu and ^{40}Ca + ^{175}Lu. The typical energy resolution for the PSSDs was about 40 keV (full width at half maximum, FWHM) for 6.5- to 10.5-MeV α particles. The vertical position of each event was determined by the resistive charge division method and the position resolution of each strip was better than 1.5 mm (FWHM) for the events with deposited energies larger than 5 MeV. The identification of rare activities of interest was performed using the spatial and time correlations between the implants and subsequent α decays.

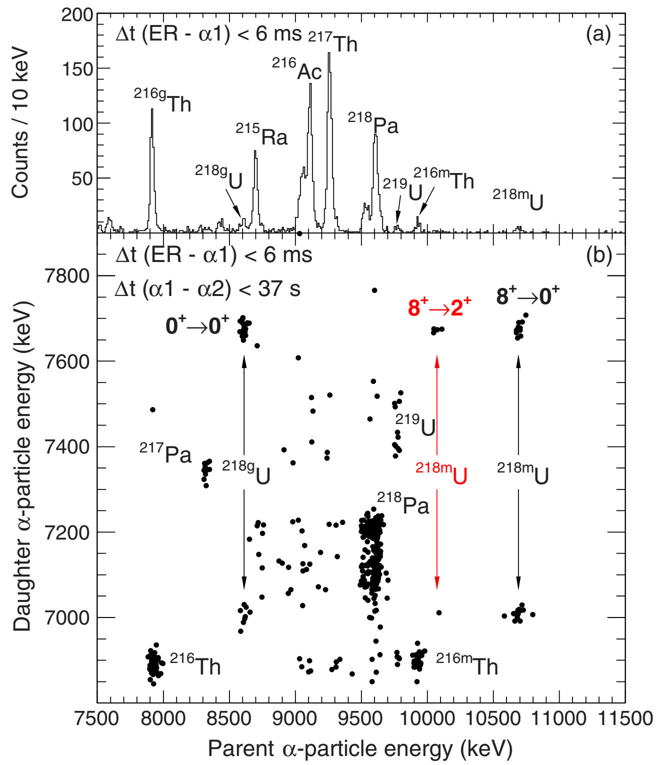

FIG. 1. (a) Energy spectrum for α particles following implanted residues within a time window of 6 ms measured in the ^{40}Ar + ^{182}W reaction. (b) Two-dimensional plot of parent and daughter α-particle energies for correlated ER-α1-α2 events measured in the PSSDs. The searching time windows were 6 ms for the ER-α1 pair and 37 s for the α1-α2 pair. The newly observed $8^+ \rightarrow 2^+$ α decays of ^{218m}U are marked with red arrows.

III. RESULTS AND DISCUSSION

A. Isotope ^{218}U

The isotope ^{218}U, as the heaviest even-even $N = 126$ isotone known experimentally to date, was first synthesized by Andreyev *et al.* [7] using the complete fusion reaction ^{27}Al + ^{197}Au. Based on four correlated α-decay chains, the α-particle energy and the half-life of ^{218}U were determined to be $E_\alpha = 8625(25)$ keV and $T_{1/2} = 1.5^{+7.3}_{-0.7}$ ms, respectively. Later, the α-decay properties of ^{218}U were reinvestigated in Refs. [8,9]. Two α-decaying states, with $E_\alpha = 8612(9)$ keV and $T_{1/2} = 0.51^{+0.17}_{-0.10}$ ms for the ground state and $E_\alpha = 10678(17)$ keV and $T_{1/2} = 0.56^{+0.26}_{-0.14}$ ms for an isomeric 8^+ state, were identified.

Figure 1(a) shows the energy spectrum measured by the PSSDs in the reaction ^{40}Ar + ^{182}W, in which only α particles following the ERs within a time window of 6 ms are included to focus on the decay of short-lived ^{218g,m}U. Several α-decay peaks from Ra, Ac, Th, Pa, and U isotopes produced in charged-particle and/or neutron evaporation channels are

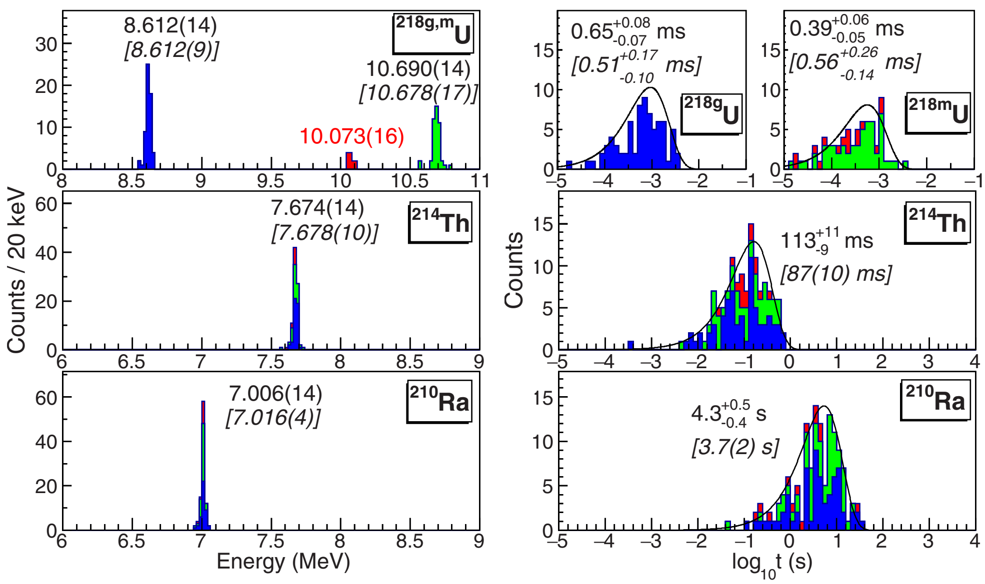

FIG. 2. The α-particle energy and the decay-time distributions of the correlated α-decay chains stemming from ^{218g}U (blue) and ^{218m}U (green) observed in the present work. The newly observed α-decay chains of ^{218m}U are marked in red. Note that only the α events with their full energy deposited in the PSSDs are shown in the energy spectra. The italic values in brackets are taken from Refs. [8,9,18,19] as comparison. The solid curves in the decay-time distributions are drawn by using the deduced mean lifetimes for ^{218g,m}U and the literature values for ^{214}Th and ^{210}Ra.

identified. In Fig. 1(b), a two-dimensional scatter plot showing the correlation between the parent and the daughter α particles is presented. The searching time windows were set to be $\Delta t(\mathrm{ER}\text{-}\alpha 1) < 6$ ms and $\Delta t(\alpha 1\text{-}\alpha 2) < 37$ s, respectively. From the plot, the α-decay correlations originated from known Th, Pa, and U isotopes are clearly identified based on their tabulated α-decay properties [19]. The correlations assigned to the decay of ^{218g,m}U are indicated by arrows.

In order to reliably identify the α decay of ^{218}U, a search for decay chains with three consecutive α decays (ER-α1-α2-α3) was performed in both runs. Figure 2 displays the measured α-particle energy and the decay-time distributions of the correlated α-decay chains stemming from ^{218g}U and ^{218m}U. Good agreement can be found between the present results and the literature data [8,9,18,19]. Forty-one and 35 decay chains were assigned to the g.s. to g.s. decay of ^{218}U in the ^{40}Ar + ^{182}W and ^{40}Ca + ^{184}W reactions, respectively. By combining the two experimental data sets, the α-particle energy and the half-life were determined to be $E_\alpha = 8612(14)$ keV and $T_{1/2} = 0.65^{+0.08}_{-0.07}$ ms for ^{218g}U, which were reported in our previous work [12]. The half-lives were extracted using the maximum likelihood method described in Ref. [21] and the α-particle energies were obtained as the arithmetic mean of the measured individual events.

Thirty-eight and 19 decay chains were attributed to the isomeric 8^+ state of ^{218}U decaying to the ground state of ^{214}Th in the ^{40}Ar + ^{182}W and ^{40}Ca + ^{184}W reactions, respectively. An α-particle energy of 10 690(14) keV and a half-life of $0.39^{+0.06}_{-0.05}$ ms were deduced, which are in good agreement with the literature data [8,9] but the present uncertainty of the half-life value is improved significantly.

Importantly, 16 new correlated decay chains (nine in the ^{40}Ar + ^{182}W reaction and seven in the ^{40}Ca + ^{184}W reaction) were observed and attributed to the ^{218m}U decaying to the known excited state of ^{214}Th. The parent activity with E_α = 10 073(16) keV and $T_{1/2} = 0.23^{+0.08}_{-0.05}$ ms was identified to be followed by a second decay with $E_\alpha = 7668(16)$ keV and $T_{1/2} = 92^{+32}_{-19}$ ms and by a third decay with $E_\alpha = 7002(16)$ keV and $T_{1/2} = 3.0^{+1.2}_{-0.7}$ s. The second decay in the chains is associated with ^{214}Th, for which the α-decay properties of E_α = 7678(10) keV and $T_{1/2} = 87(10)$ ms [22,23] were reported. The third decay in the chains can be recognized as belonging to ^{210}Ra, for which decay properties of $E_\alpha = 7016(4)$ keV and $T_{1/2} = 3.7(2)$ s were reported in Refs. [23,24]. Therefore, these decay chains are identified to originate from ^{218m}U.

The new α decays of ^{218m}U were assigned as $8^+ \rightarrow 2^+$ (^{214}Th). The branching ratios were determined to be 78(5)% and 22(5)% for $8^+ \rightarrow 0^+$ and $8^+ \rightarrow 2^+$ decays, respectively. The excitation energy for the 2^+ state of ^{214}Th was determined to be 629(22) keV according to the Q_α differences between $8^+ \rightarrow 0^+$ and $8^+ \rightarrow 2^+$ decays. This is in good agreement with the value of 623(1) keV determined previously in the ER-γ-α delayed coincidence measurement [25]. In the previous study of Ref. [9], two correlated α-decay chains were suspected as the $8^+ \rightarrow 2^+$ decay of ^{218m}U. The activity with E_α = 10 083 keV and $T_{1/2} = 0.27$ ms was followed by a decay with $E_\alpha = 7016$ keV and $T_{1/2} = 6.0$ s, which were consistent with ^{210}Ra. The authors of Ref. [9] assumed that in these two chains the second (^{214}Th) decays were lost (α particles escaping the detector), and thus the first decays represent the α-decay branch from ^{218m}U to the 2^+ state of ^{214}Th. In the present work, the α-decay properties of the $8^+ \rightarrow 2^+$ decay of ^{218m}U is confirmed undoubtedly.

The α-decay scheme for ^{218}U proposed from the present work as well as ^{214}Ra and ^{216}Th [10,11] is shown in Fig. 3. The α-decay hindrance factors (HF) were calculated using

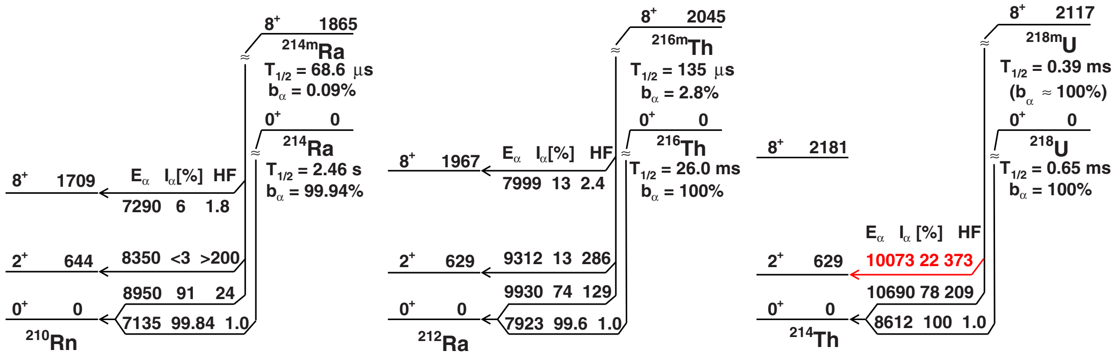

FIG. 3. α-decay schemes of the $N = 126$ even-even isotones. Values are taken from Refs. [10,11] and the present work. The α-decay hindrance factors (HF) were calculated according to Ref. [20]. Newly observed α decay from ^{218m}U to the 2^+ state of ^{214}Th is marked with a red arrow.

the relation HF $= \delta^2_{gs}/\delta^2_{ex}$, where δ^2_{gs} and δ^2_{ex} are the reduced α decay widths of the g.s. to g.s. α decays and the decays from the excited states, respectively [20]. The HF for the new α decay from ^{218m}U is determined to be 373, assuming the 100% α-decay branch of the 8^+ state. The α-decay properties of ^{214}Ra, ^{216}Th, and ^{218}U show a regular pattern in which the new data for ^{218}U fit well. It is notable that the HFs for corresponding transitions are increasing towards the higher Z nuclei, which implies an enhanced structural hindrance.

B. Isotope ^{216}U

The isotope ^{216}U was synthesized by using the reaction ^{180}W(^{40}Ar, $4n$)^{216}U in our previous work [5]. We identified two α-decaying states; the one is the ground state decaying with $E_\alpha = 8384(30)$ keV and $T_{1/2} = 4.72^{+4.72}_{-1.57}$ ms, and the other is the 8^+ isomeric state with $E_\alpha = 10\,582(30)$ keV and $T_{1/2} = 0.74^{+1.34}_{-0.29}$ ms. In the work of Ref. [4], six decay chains of ^{216}U were observed using the complete fusion reactions ^{82}Kr + ^{136,137}Ba. In addition, one α-decay chain of ^{216}U was also identified in deep inelastic multinucleon transfer reactions of ^{48}Ca + ^{248}Cm [6]. However, only a few α-decay chains of ^{216}U were observed in each study, resulting in a relatively large uncertainty of decay half-lives.

In the present study, the α-decay properties of ^{216}U were reinvestigated using the reaction ^{180}W(^{40}Ar, $4n$)^{216}U as in our previous work. An energy spectrum for the α-decay events following the ERs and a two-dimensional plot for the α-particle energy correlation between parent and daughter nuclei are displayed in Figs. 4(a) and 4(b), respectively. The Pa, Th, and Ac isotopes were produced from charged-particle evaporation channels. The pure neutron evaporation channels yield ^{216}U, ^{217}U, and ^{218}U. Here, it should be noted that the isotopes ^{218}U and ^{218}Pa were produced in the irradiation of ^{182}W with ^{40}Ar projectiles due to the impurity (8.5% ^{182}W) of our targets.

Figure 5 shows the measured α-particle energy and the decay-time distributions of the correlated α-decay chains stemming from ^{216g}U and ^{216m}U. Thirteen α-decay chains of the type ER-α1-α2-α3-(α4) were observed and assigned to the g.s. to g.s. decay of ^{216}U. Based on these chains, the α-particle energy and the half-life were determined to be $E_\alpha = 8374(17)$ keV and $T_{1/2} = 1.28^{+0.49}_{-0.28}$ ms for ^{216g}U, which were reported in our previous work [12]. Nineteen α-decay chains were attributed to the isomeric 8^+ state of ^{216}U. A half-life of $0.89^{+0.27}_{-0.17}$ ms and an α-particle energy of 10 539(16) keV were deduced for this isomer. By combing the decay-time data from the present study and from Refs. [4–6], the averaged

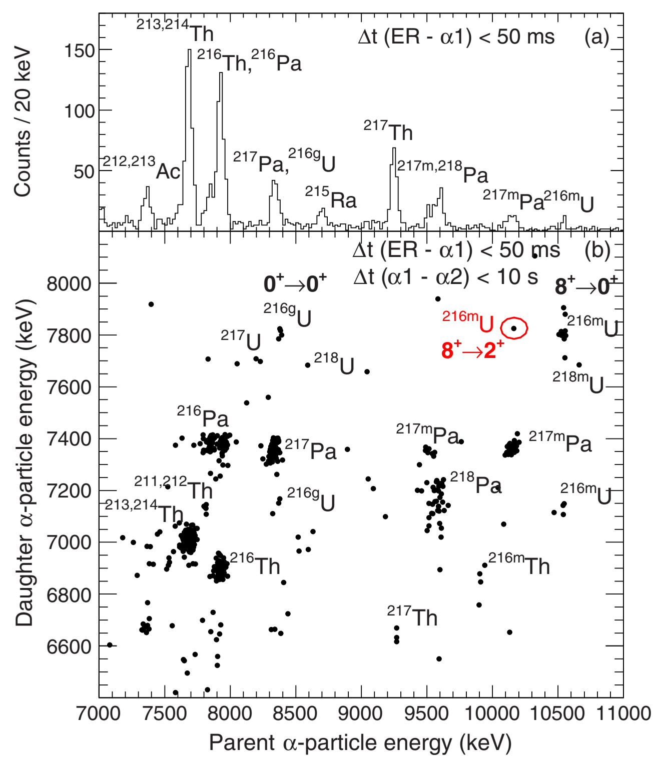

FIG. 4. (a) Energy spectrum for α particles following implanted residues within a time window of 50 ms measured in the ^{40}Ar + ^{180}W reaction. (b) Two-dimensional plot of parent and daughter α-particle energies for correlated ER-α1-α2 events measured in the PSSDs. The searching time windows were 50 ms for the ER-α1 pair and 10 s for the α1-α2 pair. The newly observed $8^+ \rightarrow 2^+$ α decay of ^{216m}U is labeled by a red ellipse.

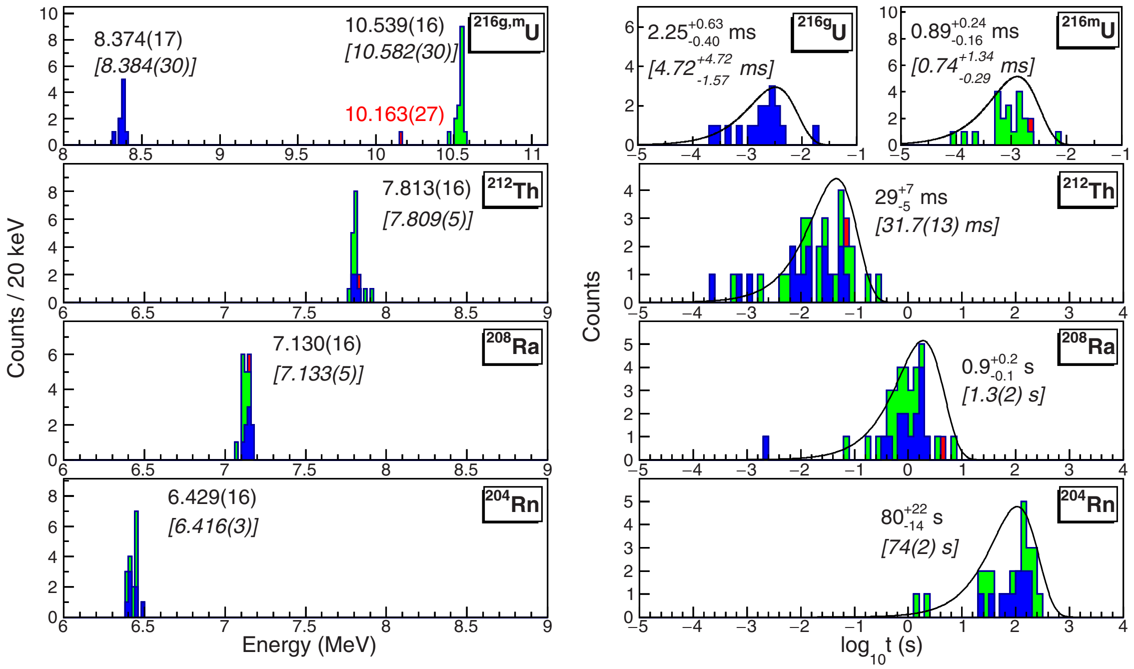

FIG. 5. Measured α-particle energy and decay-time distributions of the correlated α-decay events stemming from ^{216g}U (blue) and ^{216m}U (green). The newly observed α-decay chain of ^{216m}U is marked in red. Note that only the α events with their full energy deposited in the PSSDs are shown in the energy spectra. The half-lives of ^{216g}U and ^{216m}U are deduced by combining all the decay-time data from the present work and from Refs. [4–6]. The italic values in brackets are taken from Refs. [5,19,26,27]. The solid curves in the decay-time distributions are drawn by using the deduced mean lifetimes for ^{216g,m}U and the literature values for ^{212}Th, ^{208}Ra, and ^{204}Rn.

half-lives were deduced to be $T_{1/2} = 2.25^{+0.63}_{-0.40}$ ms for ^{216g}U and $T_{1/2} = 0.89^{+0.24}_{-0.16}$ ms for ^{216m}U, respectively.

One ER-α1-α2-α3 correlation labeled by the red ellipse in Fig. 4 is regarded as the α decay from ^{216m}U into the low-lying excited state of the daughter nucleus ^{212}Th. In Fig. 5, the measured decay properties for each chain member are drawn in red. The parent activity with $E_\alpha = 10\,163(27)$ keV and a decay time of 2.24 ms was identified to be followed by a daughter decay with $E_\alpha = 7825(27)$ keV and a decay time of 64 ms. The daughter activity can be attributed to ^{212}Th, for which decay properties of $E_\alpha = 7809(5)$ keV and $T_{1/2} = 31.7(13)$ ms [26] were reported to be originated from the ground state of ^{212}Th. The granddaughter decay with $E_\alpha = 7141(27)$ keV and a decay time of 1.1 s in the chain can be associated with ^{208}Ra, whose reported α-decay properties are $E_\alpha = 7133(5)$ keV and $T_{1/2} = 1.3(2)$ s [19]. The possibility that this observed decay chain arose from a random correlation of unrelated events was estimated to be less than 1×10^{-14} using the method described in Ref. [21]. Thus, this decay chain can be considered to be a real correlation. Because the measured energy of 10 163 keV is higher than the α energy of g.s. to g.s. and the decay time of 2.24 ms is compatible with the known half-life of ^{216m}U (0.89 ms), this decay chain was assigned to the α decay of the isomeric 8^+ state decaying to the low-lying excited state of ^{212}Th.

Due to the similar level structure and α-decay properties between ^{216}U and neighboring even-even nuclei [5,9,11], we tentatively assign the 10 163-keV α line as the decay from the isomeric 8^+ state of ^{216}U to the previously unknown 2^+ state in the daughter nucleus ^{212}Th. The proposed α-decay scheme of ^{216}U is illustrated in Fig. 6. The 2^+ state energy in ^{212}Th is determined to be 383(32) keV in the present work for the first time. For the lighter $N = 122$ isotones the first 2^+ state energies of 899, 701, 636, and 604 keV are reported for ^{204}Pb, ^{206}Po, ^{208}Rn, and ^{210}Ra, respectively [19]. Our value follows the trend of decreasing energies of the 2^+ states with increasing mass number for the $N = 122$ isotones. However, poor statistics brought large uncertainty on the

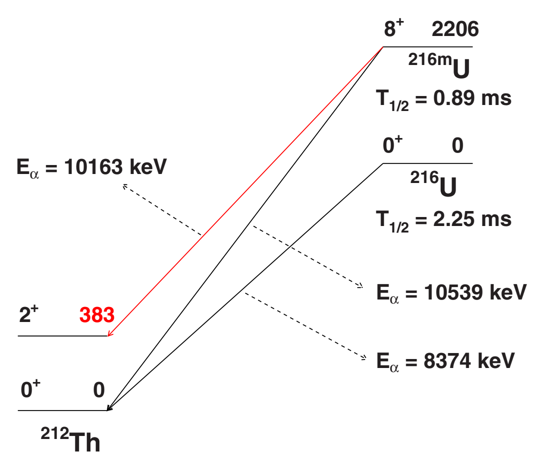

FIG. 6. Proposed α-decay scheme of ^{216}U. The newly observed α decay from ^{216m}U to the 2^+ state of ^{212}Th is marked with red.

excitation energy. Further investigations with more statistics are needed.

IV. SUMMARY

In conclusion, the extremely neutron-deficient even-even uranium isotopes ^{216,218}U were produced using the complete-fusion reactions of ^{182}W(^{40}Ar, $4n$)^{218}U, ^{184}W(^{40}Ca, $\alpha 2n$)^{218}U, and ^{180}W(^{40}Ar, $4n$)^{216}U. The α-decay properties of ^{216,218}U were measured with improved precision. A new α-decay transition with $E_\alpha = 10\,073(16)$ keV from the 8^+ isomeric state of ^{218}U to the 2^+ state in ^{214}Th was identified. The systematics in the α decay of the $N = 126$ even-even isotones ^{214}Ra, ^{216}Th, and ^{218}U have been discussed.

For ^{216}U, we have also observed a new α-decay transition with $E_\alpha = 10\,163(27)$ keV and identified it as decay from the 8^+ isomeric state of ^{216}U to the 2^+ state in ^{212}Th. The excitation energy of the 2^+ state in ^{212}Th was determined to be 383(32) keV.

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