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Ground-state bands of neutron-rich ^{236}Th and ^{242}U nuclei and implication of spherical shell closure at $N = 164$

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The ground-state bands of the neutron-rich ^{236}Th and ^{242}U nuclei were established up to spin 10 and 8, respectively, by in-beam γ-ray spectroscopy using the (^{18}O, ^{20}Ne) two-proton pickup reaction with a ^{238}U and a ^{244}Pu target. Deexcitation γ rays in ^{236}Th and ^{242}U were identified by selecting the kinetic energies of ^{20}Ne using Si ΔE-E detectors. The excitation energies of the first 2^+ states in U and Pu isotopes have local minima at $N \simeq 146$, suggesting the possibility that nuclei with $Z \simeq 92$ have a spherical shell closure of $N = 164$. Calculation using the Koura-Yamada single-particle potential gives an energy gap of 1.8 MeV at $N = 164$ for ^{256}U.

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Shell structure plays a key role in stabilizing superheavy nuclei. At the present time, however, different magic numbers are predicted by different theories and parameters included within the theories. Most macroscopic-microscopic calculations predict the magic numbers of $Z = 114$ and $N = 184$ [1], while self-consistent relativistic mean field calculations predict other magic numbers, such as $Z = 120, 126$ and $N = 172, 184$ [1,2]. Single-particle energies in heavy- and transactinide nuclei in the deformed region give important information for predicting the shell structure of spherical superheavy nuclei, because some of the single-particle states in deformed nuclei with $A \sim 250$ derive from the spherical single-particle states at $A \sim 300$. The systematic study of collective states in those deformed nuclei also gives a clue to the shell closure of spherical superheavy nuclei.

Nuclear structure for nuclei with $Z \geqslant 100$ has been recently studied by in-beam γ-ray and isomer γ-ray spectroscopy coupled to the recoil mass separator [3–6] and by α-γ spectroscopy [7–9]. These nuclei are, however, limited to the proton-rich side of the β-stability line, because they were produced by heavy-ion fusion reactions. Nuclear structure of heavy-actinide nuclei at the neutron-rich side has been scarcely studied experimentally. We have measured in-beam γ rays of neutron-rich ^{240}U, ^{246}Pu, and ^{250}Cm nuclei produced by the (^{18}O, ^{16}O) two-neutron transfer reactions [10–12]. In these measurements, the γ rays in the residual nuclei were selected by measuring outgoing ^{16}O nuclei using Si ΔE-E detectors. In this Rapid Communication, we will report the results of in-beam γ-ray study of more neutron-rich ^{236}Th and ^{242}U nuclei produced by the (^{18}O, ^{20}Ne) two-proton pickup reactions. Excited states in these nuclei have never been measured. We will discuss the possibility of the spherical shell closure of $N = 164$ through the systematics of the excitation energies of the first 2^+ states in actinide nuclei and the calculation of an accurate spherical single-particle potential.

The experiments were carried out at the JAEA-Tokai tandem accelerator facility [13,14]. As for the ^{236}Th experiment, a natural uranium target, 4 mg/cm^2 in thickness, electrodeposited on a 3 μm aluminum foil was bombarded by a 200-MeV ^{18}O beam with 0.1 particle nA. As for the ^{242}U experiment, a 98% enriched ^{244}Pu target, 0.7 mg/cm^2 in thickness, electrodeposited on a 3 μm aluminum foil was bombarded by a 162-MeV ^{18}O beam with 0.3 particle nA. The total doses of ^{18}O ions were 1.3×10^{14} and 4.4×10^{14} for the ^{236}Th and ^{242}U experiments, respectively. Residual nuclei produced by the transfer reaction stop in the target or the aluminum backing. Most excited states up to medium-spin states have lifetimes longer than the stopping time. Therefore, the γ rays emitted by these states can be measured without Doppler shifts.

Outgoing nuclei were detected with four sets of Si ΔE-E detectors of 20 mm in diameter. These Si ΔE-E detectors were placed at a distance of 5 cm from the target, at 28° to the beam axis for the ^{236}Th experiment and at 40° for the ^{242}U experiment; these angles are slightly smaller than the grazing angles. The ΔE detectors were surface-barrier Si detectors made from an ELID (ELectrolytic In-process Dressing)-grinding Si wafer. The thicknesses of the ΔE detectors were 83 μm and 75 μm for the ^{236}Th and ^{242}U experiments, respectively.

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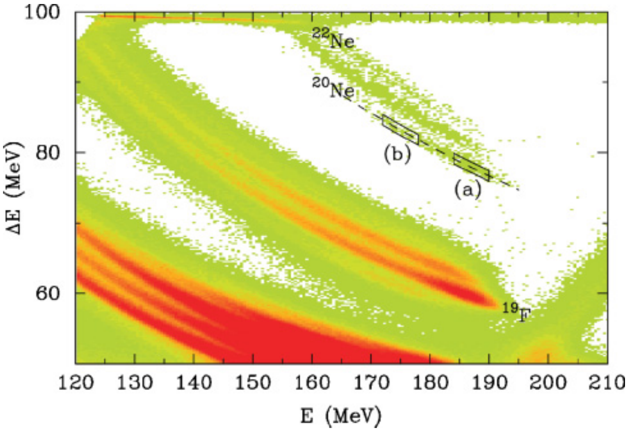

FIG. 1. (Color online) E-ΔE plot of scattered nuclei measured by a Si ΔE-E detector in the reaction of a 200-MeV ^{18}O beam with a ^{238}U target. The dashed line represents a calculated energy loss for ^{20}Ne nuclei. See the caption of Fig. 2 for the enclosed areas (a) and (b).

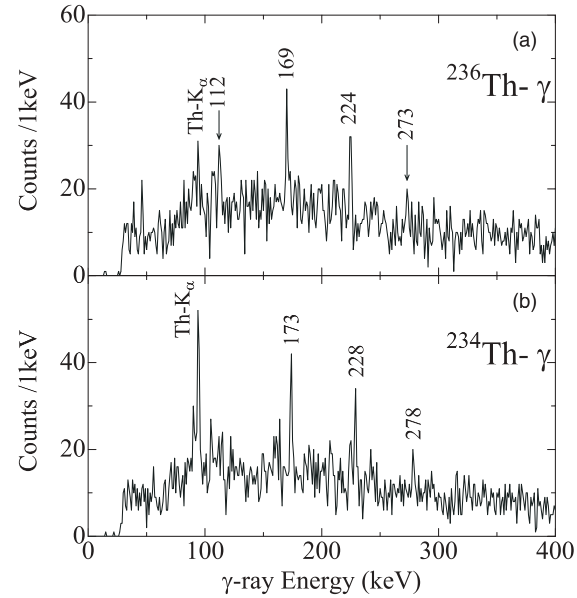

FIG. 2. γ-ray spectra obtained by setting the gates on ^{20}Ne whose kinetic energies correspond to E_x of (a) 5–11 and (b) 17–23 MeV, respectively. These gates are depicted as the regions (a) and (b) in Fig. 1, respectively. γ peaks labeled by energies in the spectra of (a) and (b) are the transitions in ^{236}Th and ^{234}Th, respectively.

Deexcitation γ rays in coincidence with the outgoing nuclei were measured by Ge detectors surrounding the target; seven Ge detectors were employed for the ^{236}Th experiment and six detectors for the ^{242}U experiment. Four Ge detectors, with 60% relative efficiency, were arranged symmetrically in the plane perpendicular to the beam axis, at a distance of 6 cm from the target. The remaining two or three Ge detectors with 30–40% relative efficiency were placed between the former Ge detectors. The absolute efficiencies of the total Ge detectors were about 3% for a 1.33-MeV γ ray.

An E-ΔE plot obtained from the ^{236}Th experiment is shown in Fig. 1. The dashed line in Fig. 1 represents a calculated energy loss for ^{20}Ne nuclei. The distribution of ^{20}Ne was distinguished from those of Ne isotopes with different mass numbers. When the ^{20}Ne nucleus is emitted in the transfer reaction, the residual nucleus is ^{236}Th. The sum of excitation energies of scattered and residual nuclei, E_x, is derived from the kinetic energy of ^{20}Ne; a lower kinetic energy of ^{20}Ne corresponds to higher E_x. The region (a) in Fig. 1 corresponds to E_x between 5 and 11 MeV. By taking coincidence with ^{20}Ne within the region (a), we have observed almost equally spaced transitions of 112, 169, 224, and 273 keV as shown in Fig. 2(a). They are the candidates for the ground-state band transitions in ^{236}Th. We have observed no distinct γ rays in coincidence with ^{20}Ne having larger kinetic energies than the region (a). Since the two-proton pickup reaction has a highly negative Q_{eff} value [15], proton transfer to excited states with large angular momentum in ^{20}Ne would be favored for satisfying matching condition of angular momentum. Thus, the γ rays in ^{236}Th may not be observed at $E_x < 5$ MeV. This fact is in contrast to the results of the (^{18}O, ^{16}O) stripping reaction where the γ rays in residual nuclei were observed in the range of E_x between 0 and 6 MeV [10–12].

To confirm the assignment of γ rays in ^{236}Th, we have examined the γ rays in coincidence with ^{20}Ne having lower kinetic energies than the region (a). Gates of ^{20}Ne were set by dividing the ^{20}Ne distribution into 6 MeV in width; this energy corresponds to a neutron separation energy of ^{236}Th. In the next gate of the region (a), the candidates for γ rays in ^{236}Th were hardly observed. In the succeeding gate, depicted as the region (b) in Fig. 1, previously identified γ rays in ^{234}Th appeared [16,17]. Figure 2(b) shows the γ-ray spectrum coincident with the region (b). Because E_x for the region (b) is higher than that for the region (a) by 12 MeV, the ^{236}Th nucleus is excited high enough to evaporate two neutrons at the region (b). This observation of γ rays in ^{234}Th suggested that the γ rays coincident with the region (a) are the deexcitation γ rays in ^{236}Th. The yield of ^{236}Th produced by the two-proton pickup reaction was two orders of magnitude smaller than that of ^{240}U produced by the two-neutron stripping reaction. The cross section of ^{236}Th was on the order of μb.

Following the same analysis, we have identified γ rays in ^{242}U produced by the ^{244}Pu(^{18}O, ^{20}Ne) reaction. Figure 3

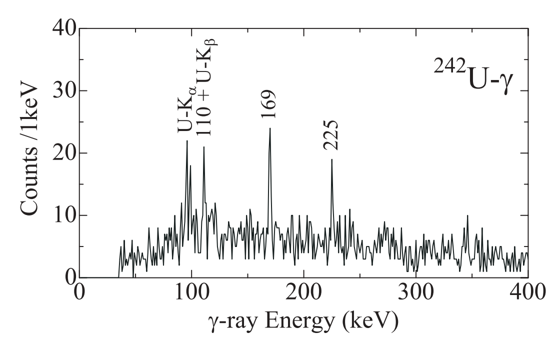

FIG. 3. γ-ray spectrum of ^{242}U measured in the reaction of a 162-MeV ^{18}O beam with a ^{244}Pu target. This spectrum was obtained by setting the gate on ^{20}Ne whose kinetic energies correspond to E_x between 5 and 11 MeV. γ peaks labeled by energies are the transitions in ^{242}U.

TABLE I. γ-ray energies and relative intensities in ^{236}Th and ^{242}U. Total internal conversion coefficients α_T for $E2$ transitions were taken from Ref. [18].

$I_i \to I_f$	^{236}Th		^{242}U	
	E_γ (keV)	$I_\gamma(1+\alpha_T)$	E_γ (keV)	$I_\gamma(1+\alpha_T)$
$2^+ \to 0^+$	(48.4(3))[a]		(47.8(3))[a]	
$4^+ \to 2^+$	111.6(5)	240(70)	110.4(6)	170(70)
$6^+ \to 4^+$	169.4(3)	100(22)	169.3(3)	100(20)
$8^+ \to 6^+$	224.0(3)	48(12)	224.7(3)	44(14)
$10^+ \to 8^+$	272.7(5)	32(14)		

[a]The energy was derived from the moment of inertia deduced from the higher levels.

shows the γ rays in coincidence with ^{20}Ne with the kinetic energies corresponding to E_x between 5 and 11 MeV. We have assigned the 110, 169, and 225 keV γ rays to deexcitation γ rays in ^{242}U. The γ-ray energies and intensities in ^{236}Th and ^{242}U are summarized in Table I. Assuming that those γ rays form the ground-state rotational bands, we have established level schemes of ^{236}Th and ^{242}U up to spin 10 and 8, respectively, as shown in Fig. 4; the $2^+ \to 0^+$ γ transitions were not observed owing to large internal conversion coefficients, $\alpha_T > 300$ [18].

The moments of inertia for the ground-state band of ^{242}U are plotted as a function of squared rotational frequency in Fig. 5. $\mathcal{I}^{(1)}$ and $\mathcal{I}^{(2)}$ represent the kinematic and dynamic moments of inertia, and ω is rotational frequency [10,12]. The $\mathcal{I}^{(1)}$ values were derived from the spins assigned above as well as the γ-ray energies (Table I). The solid line in Fig. 5 is the fit of $\mathcal{I}^{(1)}$ to $J_0 + \omega^2 J_1$, where J_0 and J_1 are constants. The $\mathcal{I}^{(2)}$ values calculated by $J_0 + 3\omega^2 J_1$, drawn as the dashed line in Fig. 5, are in good agreement with the experimental ones. This fact suggests that the spin assignments of the levels in ^{242}U are correct. From the J_0 and J_1 values, the 2^+ energies were deduced to be 48.4(3) and 47.8(3) keV for ^{236}Th and ^{242}U, respectively. These 2^+ energies are very precise, which would be difficult to be attained by other measurements.

The excitation energies of the 2^+ states in the ground-state bands, E_{2^+}, are plotted versus neutron number in Fig. 6 for even-even actinide nuclei whose excitation energies are known precisely [3,10–12,19–23]; five data for

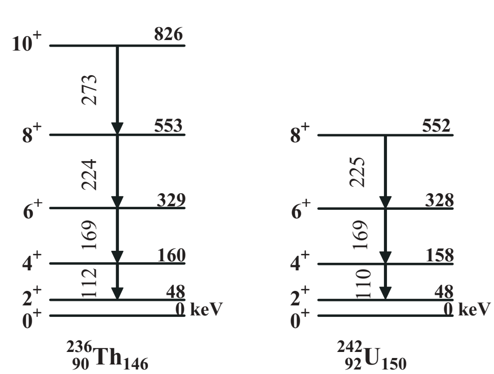

FIG. 4. Level schemes of ^{236}Th and ^{242}U. The γ-ray and level energies are in units of keV.

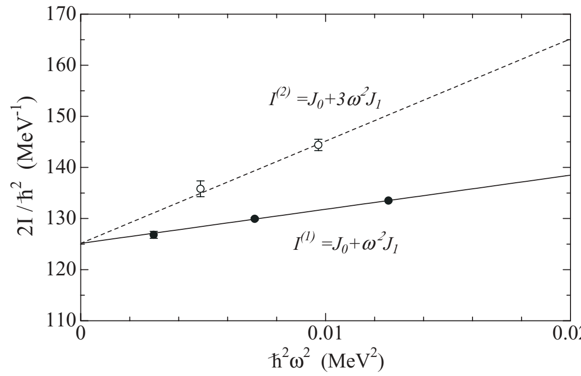

FIG. 5. Plot of moments of inertia for the ground-state band of ^{242}U versus squared rotational frequency ω^2. The closed and open circles represent the kinematic moments of inertia $\mathcal{I}^{(1)}$ and the dynamic moments of inertia $\mathcal{I}^{(2)}$, respectively. The solid line is the fit of $\mathcal{I}^{(1)}$ to $J_0 + \omega^2 J_1$. The dashed line is calculated by $J_0 + 3\omega^2 J_1$, using $J_0 = 62.6(2)\hbar^2$ MeV^{-1} and $J_1 = 334(23)\hbar^4$ MeV^{-3} obtained from the fit of $\mathcal{I}^{(1)}$.

^{236}Th$_{146}$, ^{240,242}U$_{148,150}$, ^{246}Pu$_{152}$, and ^{250}Cm$_{154}$ were obtained from a series of our studies. In this systematics, the E_{2^+} values of Th and U isotopes have dips at $N = 142$. Furthermore, the E_{2^+} values of No isotopes decrease at $N = 152$. These sharp decreases of E_{2^+} are related to the deformed shell gaps. At the deformed shell gap, the pairing energy gap becomes smaller, resulting in a larger moment of inertia. In fact, there are deformed shell gaps at $N = 142$ [24,25] and 152 [25–27]. In Ref. [12], we have discussed the relation between the moments of inertia of Cm isotopes and the deformed shell gap at $N = 152$. The disappearance of this gap for Pu isotopes will also be discussed in another paper [11]. By disregarding the dips at the deformed shell gaps, we have found that the E_{2^+} values vary smoothly and have local minima at $N \simeq 146$ for U and Pu isotopes, and possibly for Th and Cm isotopes.

From a simple perspective on nuclear structure, we can consider that nuclei in the middle of the spherical shell closures have maximum collectivity, and thus, the excitation energies of the first 2^+ states may have local minima there. In fact,

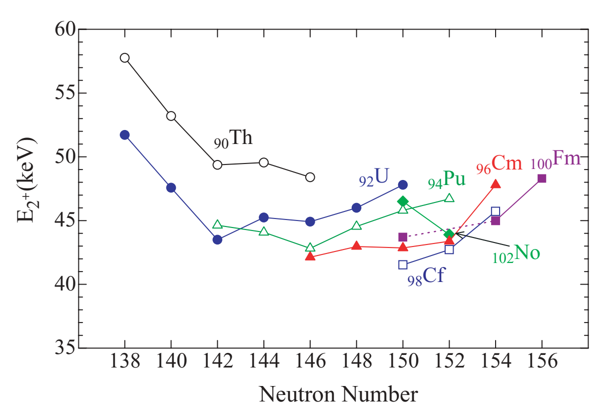

FIG. 6. (Color online) Systematics of the first 2^+ energies of even-even actinide nuclei. The data for ^{250}Fm and ^{254}Fm are connected by a dotted line because there are no accurate measurements for ^{252}Fm$_{152}$.

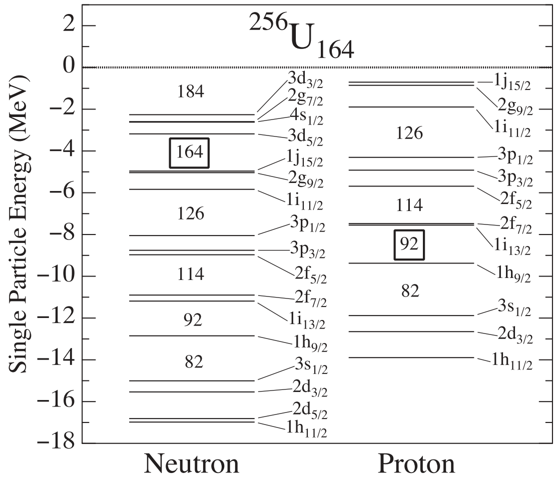

FIG. 7. Single-particle levels calculated by the KY potential for $^{256}U_{164}$.

this perspective is broadly true in other deformed regions. In the actinide region, one side of the spherical shell closure of neutrons is $N = 126$. Therefore, the other side of the spherical shell closure becomes $N = 166$ by regarding $N = 146$ as the middle point. Among the single-particle levels lying around $N = 166$, it is very plausible that there is a shell gap at $N = 164$ between the $1j_{15/2}$ orbital and the $3d_{5/2}$ or $2g_{7/2}$ orbitals. This gap is similar to the $N, Z = 50$ gap lying between the $1g_{9/2}$ orbital and the $2d_{5/2}$ or $1g_{7/2}$ orbitals. Furthermore, protons may have a spherical subshell closure of $Z = 92$, where the $h_{9/2}$ orbital is fully occupied. Therefore, the ^{256}U nucleus with $Z = 92$ and $N = 164$ may have a doubly shell closure.

The macroscopic-microscopic calculation using a multidimensional deformation space [28] reproduced and predicted the E_{2^+} values of heavy- and transactinide nuclei very well. The E_{2^+} values of ^{232,234,236}Th were calculated as 47.3, 46.9, and 46.5 keV (experimental energies are 49.4, 49.6, and 48.4 keV), respectively, and those of ^{238,240,242}U were calculated as 43.7, 44.9, and 48.0 keV (experimental energies are 44.9, 46.0, and 47.8 keV), respectively. Their calculation also well reproduced the correlation between the E_{2^+} values and the deformed shell gap at $N = 152$. They, however, did not mention nuclear structure of more neutron-rich nuclei around ^{256}U.

Calculation using the Koura-Yamada (KY) single-particle potential [29] shows that shell gaps appear at $N = 164$ and $Z = 92$ for ^{256}U, as shown in Fig. 7. This potential is an extension of Woods-Saxon potential including two new parameters modifying the surface structure of the potential. This potential is expressed as a smooth function of Z and N, and the potential parameters are fixed by comparison with single-particle levels in the vicinities of 15 doubly magic or magic-submagic nuclei ranging from ^{4}He to ^{208}Pb. Because this potential is very accurate and applicable to a wide region of nuclides [29], the KY potential is expected to have more predictive power than a simple Woods-Saxon potential. The KY potential gives a shell gap of 1.8 MeV at $N = 164$ and 1.8 MeV at $Z = 92$ for ^{256}U. These energy gaps are comparable size to those calculated by the KY potential for an expected doubly-magic superheavy nucleus of ^{298}114 (2.5 MeV at $N = 184$ and 1.6 MeV at $Z = 114$), and are about half of those for a typical doubly magic nucleus of ^{208}Pb. In the calculation of the KTUY mass formula [30] using the KY potential, it is also predicted that nuclei around ^{256}U are almost spherical. Note that the Woods-Saxon potential using 'universal parameters' [31], which is often used for the calculations of energy levels in heavy- and transactinide nuclei [5,28,32], gives a smaller energy gap of 1.1 MeV at $N = 164$ for ^{256}U than the KY potential.

In conclusion, we have measured deexcitation γ rays in the neutron-rich ^{236}Th and ^{242}U nuclei for the first time. These nuclei were produced by the (^{18}O, ^{20}Ne) two-proton pickup reactions, and their γ rays were identified by taking coincidence with ^{20}Ne nuclei and selecting their kinetic energies, using Si ΔE-E detectors. We have found that the excitation energies of the first 2^+ states in U and Pu isotopes have local minima at $N \simeq 146$, disregarding the E_{2^+} value of ^{234}U having the deformed shell closure of $N = 142$. A spherical shell closure of $N = 164$ is expected for nuclei with $Z \simeq 92$, which would also affect the prediction of the path of the r-process for heavy nuclei.

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