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EC and α decays of ^{235}Am

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Abstract. EC and α decays of ^{235}Am have been studied using a gas-jet coupled on-line isotope separator. Excited states in ^{235}Pu have been established for the first time by means of γ-ray spectroscopy following the EC decay of ^{235}Am. The deduced $\log ft$ value suggests that the ground state of ^{235}Am should have the $\pi 5/2^-[523]$ configuration. The α-γ coincidence result has revealed that the $\pi 5/2^-[523]$ state in ^{231}Np populated by the favored α transition of ^{235}Am is located at < 15 keV, which allows us to precisely determine the Q_α value of ^{235}Am.

PACS. 23.20.Lv γ transitions and level energies – 23.40.-s β decay; double β decay; electron and muon capture – 23.60.+e α decay – 27.90.+b $220 \leq A$

1 Introduction

Gamma-ray spectroscopy of heavy actinide nuclei enables us to establish level structure and Nilsson single-particle states which provide detailed information on shell structure in the region of heavy and superheavy nuclei. The decay of neutron-deficient Am isotopes has been studied scarcely. These nuclei predominantly decay by electron capture (EC) and their α-decay branching ratios are extremely small. Although EC decays of $^{237-240}$Am have been studied in detail using an off-line mass separation technique coupled to a chemical purification of Am atoms owing to their long half-lives of > 1 h [1–4], no γ transition has been observed in the decay of more neutron-deficient Am isotopes, because their half-lives are shorter than $\sim$10 min, which makes it difficult to prepare low-contaminated α/γ sources. To study these nuclei, we have developed a gas-jet coupled on-line isotope separator (ISOL) [5,6]. Extremely pure sources from the ISOL enable us to study EC decays of short-lived actinide nuclei by means of γ-ray spectroscopy and to observe very weak α transitions with unambiguous mass identification.

The nucleus ^{235}Am was identified for the first time by Guo *et al.* [7]. They produced ^{235}Am by the ^{238}Pu$(p, 4n)$ reaction, and observed a growth and decay of the daughter nucleus ^{235}Pu in chemically purified Am fractions. Only the half-life value of 15(5) min was derived for the EC decay of ^{235}Am. There has been no experimental information on excited states and γ transitions in ^{235}Pu. The α-decay of ^{235}Am was identified in our previous experiments [8] using the ISOL. The α energy of 6457(14) keV, its intensity 0.40(5)%, and the half-life value of 10.3(6) min were determined.

In this paper, we present the γ-ray spectroscopic studies for the EC and α decays of ^{235}Am, and discuss excitation energies of Nilsson single-particle states in ^{235}Pu and ^{231}Np as well as the ground-state configuration of ^{235}Am. The Q_α value of ^{235}Am is determined more accurately than that of ref. [8] through an additional analysis of the α-γ coincidence data. Part of this work has been published in ref. [9].

2 Experiments

The nucleus ^{235}Am was produced by the ^{233}U(^{6}Li, $4n$)^{235}Am reaction at the JAERI tandem accelerator facility. A stack of twenty-one ^{233}U targets set in a multiple-target chamber with 5 mm spacings was bombarded with

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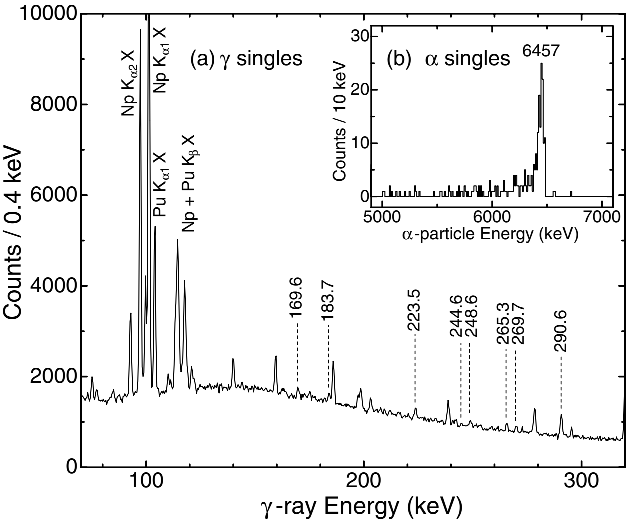

Fig. 1. (a) Gamma-ray singles spectrum for the mass-235 fraction. Gamma-rays associated with the EC decay of ^{235}Am are indicated by their energies in keV. Unlabelled γ lines mostly originate from background γ-rays. (b) Alpha singles spectrum.

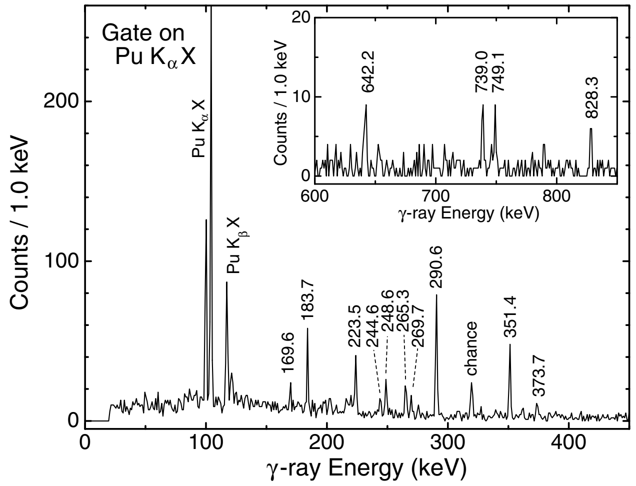

Fig. 2. Gamma-ray spectrum in coincidence with Pu K_α X-rays. The inset shows a high-energy portion of the spectrum.

Table 1. Energies, relative intensities, and coincidence relationships of γ-rays in the EC decay of ^{235}Am.

Energy (keV)	Relative intensity	Coincident γ-rays (keV)
Pu $K_{\alpha1}$ X	240(50)	All the γ-rays listed in this table.
169.6(2)$^{(a)}$	13(4)	
183.7(2)	20(6)	351.4, 642.2
223.5(2)	42(9)	269.7, 373.7
244.6(2)	13(6)	290.6
248.6(2)	19(8)	828.3
265.3(2)	35(8)	269.7
269.7(2)	38(11)	223.5, 265.3
290.6(2)	100(14)	244.6, 739.0, 828.3
351.4(2)	30(9)	183.7
373.7(2)	33(12)	223.5, 265.3
642.2(2)	20(6)	183.7
739.0(2)	36(11)	290.6
749.1(2)$^{(a)}$	36(11)	
828.3(2)	27(8)	290.6

(^{a}) Not placed in the decay scheme.

a ^{6}Li beam of 450 particle-nA intensity. Each target was electrodeposited on a 0.8 mg/cm^2 thick aluminum foil with an effective target thickness of about 100 μg/cm^2. The energy of the ^{6}Li beam was 34–42 MeV on targets. Reaction products recoiling out of the targets were stopped in He gas loaded with PbI$_2$ clusters, and transported into an ion source of the ISOL by a gas-jet stream through an 8 m long capillary. Atoms ionized in the surface-ionization–type thermal ion source were accelerated with 30 kV and mass-separated with a resolution of $M/\Delta M \sim 800$. The overall efficiency of this ISOL system including a gas-jet transport efficiency and ionization for Am atoms was measured to be 0.3% through the observation of ^{237}Am produced in the ^{235}U(^{6}Li, $4n$) reaction [10].

The separated ions were continuously implanted into a Si PIN photodiode detector (Hamamatsu S3590-06, 9×9 mm^2) for the α-particle detection which was tilted 45° with respect to the ion beam axis and contained in a thin vacuum chamber having a 0.4 mm thick beryllium window on one side and 0.5 mm thick aluminum on the other side. A short coaxial Ge detector (ORTEC LOAX) was placed behind the Be window to detect low-energy γ-rays down to ∼10 keV, and a 35% n-type Ge detector (ORTEC GAMMA-X) placed on the Al side detected γ-rays from the implanted nuclei through the Si wafer and its mount. The distance between the implanted source and the endcap surface of each Ge detector was 10 mm. Alpha singles, γ-ray singles, and α-γ and γ-γ coincidence events were accumulated during 82 h in an event-by-event mode.

The energy calibration of the Si detector was performed before and after the on-line experiment using mass-separated ^{221}Fr and its α-decay daughters ^{217}At and ^{213}Po; the ^{221}Fr nuclei recoiling out of an ^{225}Ac α source were transported into the ion source by the He jet and then implanted into the Si detector by the present ISOL system. Gamma-ray energy was calibrated using a ^{152}Eu source and also using background γ lines in on-line spectra. The efficiency of the Ge detectors was measured using a mixed γ-ray standard source.

3 Results

Figure 1(a) shows a γ-ray singles spectrum for the mass-235 fraction. Np and Pu K X-rays originating from the EC decay of ^{235}Pu and ^{235}Am were clearly observed. Fourteen γ transitions were attributed to the EC decay of ^{235}Am on the basis of coincidences with both Pu K X- and L X-rays and coincidences with each other. The spectrum coincident with Pu K_α X-rays is shown in fig. 2. These γ-rays are present in the singles spectrum shown in fig. 1(a) but not seen in the background spectrum and in the adjacent mass fractions. Energies, relative intensities,

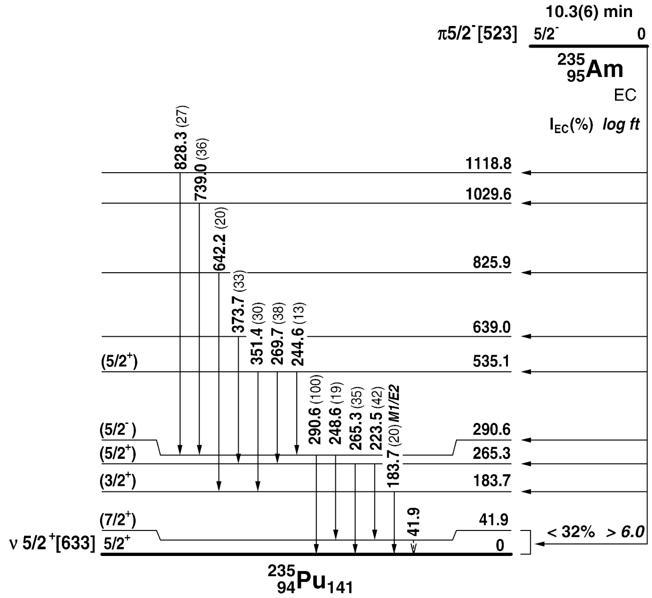

Fig. 3. Proposed decay scheme of ^{235}Am. The 41.9 keV γ-transition has not been observed because of its large internal conversion coefficient.

and coincidence relationships for these γ-rays are summarized in table 1. The energies were determined using coincidence spectra, while the intensities were deduced from both the singles and coincidence spectra. The large uncertainties in the intensities are due to low statistics and poor peak-to-background ratios. Figure 3 shows a proposed decay scheme of ^{235}Am established on the basis of the coincidence relationships. The transition depopulating the 41.9 keV first-excited state was not observed probably due to its large internal conversion coefficient.

The K internal conversion coefficient of $\alpha_K = 3.3(13)$ for the 183.7 keV transition was determined from the intensity ratio between the 183.7 keV γ-rays and Pu K_α X-rays in coincidence with the 351.4 keV γ-rays. The contribution of electron capture X-rays was subtracted in this analysis. Theoretical α_K values of the $E1$, $M1$, and $E2$ transitions are 0.098, 4.0, and 0.17, respectively [11]. The experimental α_K value is consistent with the $M1/E2$ assignment for the 183.7 keV transition. Conversion coefficients of other transitions were not determined because of low statistics.

Figure 1(b) shows an α singles spectrum measured in the present experiment. The 6457 keV α peak of ^{235}Am was clearly observed. The deduced energy of 6457(12) keV agreed very well with the previous value [8]. Although a total of 230 α-particles were detected at 6457 keV, no α-coincident γ-ray peak was observed. This result suggests that the level fed by the 6457 keV α transition is either the ground state or a low-lying state depopulated by γ transitions with large internal conversion coefficients [8].

4 Discussions

In this section, first we evaluate the spin-parity of the ground state of ^{235}Am on the basis of the deduced $\log ft$ value of the EC transition to the ground state of ^{235}Pu. Then, the level energy of ^{231}Np populated by the α-decay of ^{235}Am is estimated from the α-γ coincidence result, and the Q_α value of ^{235}Am is deduced. Finally we suggest tentative spin-parity and Nilsson orbital assignments for the excited states in ^{235}Pu on the basis of the γ-ray branching ratios compared with those of other $N = 141$ isotones.

EC transition intensities from ^{235}Am to the levels in ^{235}Pu are not determined explicitly because multipolarities of the observed γ transitions, *i.e.*, exact values of total internal conversion coefficients, are not known. In order to estimate the upper limit of the EC transition intensity to the ground state, we compare the observed Pu K X-ray intensity with the calculated one based on the proposed decay scheme. Multipolarities of all the γ transitions are assumed as $E1$ except for the known $M1/E2$ transition of 183.7 keV because the $E1$ assumption gives the smallest Pu K X-ray intensity. Theoretical K conversion coefficients [11], K fluorescence yield, and K electron capture ratios [12] are used in the calculations, and the coincidence summing effect is taken into account to deduce the K X-ray intensity. Note that the intensity of the β^+-decay is more than two orders of magnitude smaller than that of the EC decay [13]. The excess of the observed Pu K X-ray intensity against the calculated one indicates the existence of missing γ and EC transitions in the decay scheme including the EC transitions to the ground state and the 41.9 keV level. Assuming that the excess of the Pu K X-ray intensity originates only from the EC transition near to the ground state, the missing EC transition intensity is deduced to be 20(12)%, which leads to an upper limit of $< 32\%$ for the EC transition to the ground state and the 41.9 keV level. However, it should be noted that this $E1$ assumption cannot reproduce the Pu K X-ray intensity observed in coincidence with the Pu K_α X-rays shown in fig. 2. This fact suggests that at least some intense γ transitions should have an $M1$ multipolarity, which leads to a much smaller intensity of this EC transition.

$\log ft$ values are calculated using the $\log f$ table [13] and $Q_{\mathrm{EC}} = 2430(60)$ keV obtained from the measured α-particle energy and the evaluated masses of the daughter nuclei ^{231}Np and ^{235}Pu [14]. Using the intensity of $< 32\%$, the $\log ft$ value of > 6.0 is derived for the EC transition to the ground state and the 41.9 keV level.

It is known that the 95th proton of the ground state of $^{237-243}$Am occupies the $\pi 5/2^-[523]$ orbital, and the $\pi 5/2^+[642]$ orbital lies close to the Fermi surface; the $\pi 5/2^+[642]$ hole state is located at 187, 206, and 84 keV in ^{239,241,243}Am, respectively [12]. Other orbitals lie at higher excitation energy. For neutron-deficient Am nuclei, deformation changes are estimated to be small in not only β_2 but also β_4 [15,16]. Thus, it is expected that the structure of low-lying states would not change much, and the 95th proton of the ground state of ^{235}Am would occupy either the $\pi 5/2^-[523]$ orbital or the $\pi 5/2^+[642]$. As discussed in the following paragraph and suggested in ref. [17], the ground state of ^{235}Pu has the $\nu 5/2^+[633]$ configuration, that is, the EC transition to the ground state of

Table 2. Logft values of EC and β^- transitions between various proton and neutron orbitals expected to appear around ^{235}Am.

EC and β^- transition	Parent nuclide	log ft [12]
$\pi5/2^+[642] \leftrightarrow \nu5/2^+[633]$	^{232}Np, ^{233}Np, ^{235}Pu, ^{229}Pa, ^{231}Th	5.2, 5.3, 5.4, 5.7, 5.9
$\pi5/2^+[642] \leftrightarrow \nu5/2^+[622]$	^{241}Np, ^{245}Am, ^{239}U, ^{233}Np, ^{239}Np	5.9, 6.4, 6.5, 6.7, 6.8
$\pi5/2^+[642] \leftrightarrow \nu7/2^+[624]$	^{241}Np, ^{243}Pu, ^{245}Am, ^{239}Np	6.0, 6.3, 6.3, 7.3
$\pi5/2^+[642] \leftrightarrow \nu7/2^-[743]$	^{239}Np, ^{235}Np, ^{237}Pu	6.5, 6.8, 6.8
$\pi5/2^-[523] \leftrightarrow \nu5/2^+[633]$	^{235}Pu, ^{237}Am	6.5, 7.1
$\pi5/2^-[523] \leftrightarrow \nu5/2^+[622]$	^{241}Pu, ^{239}U, ^{240}Np, ^{237,239,240}Am, ^{238}Am	5.8, 5.9, 5.9, 6.0, 6.0, 6.0, 6.2
$\pi5/2^-[523] \leftrightarrow \nu7/2^+[624]$	^{239}Am, ^{243}Pu, ^{237}Am	5.9, 6.1, 6.9
$\pi5/2^-[523] \leftrightarrow \nu3/2^+[631]$	^{237}Am	7.5
$\pi5/2^-[523] \leftrightarrow \nu7/2^-[743]$	^{237}Pu, ^{237}Am, ^{239}Am	7.3, > 7.3, 8.7
$\pi5/2^-[523] \leftrightarrow \nu5/2^-[752]$		No data
$\pi7/2^+[633] \leftrightarrow \nu7/2^+[624]$	^{243}Pu	5.5

^{235}Pu would be either the $\pi5/2^-[523]\rightarrow\nu5/2^+[633]$ or the $\pi5/2^+[642]\rightarrow\nu5/2^+[633]$ transition. The deduced log ft value of > 6.0 strongly suggests the former assignment owing to the following discussion.

In table 2, we summarize the experimental log ft values of EC and β^- transitions between various proton and neutron orbitals [12] expected to appear around ^{235}Am. In addition, we surveyed log ft values of all the $Z = 90$–97 nuclei evaluated in ref. [12], and found that most of EC and β^- transitions show log $ft \gtrsim 5.8$ and only a few transitions show small log ft values of < 5.8, *e.g.*, the transitions between the $\pi5/2^+[642]$ and the $\nu5/2^+[633]$ orbitals and that between the $\pi7/2^+[633]$ and the $\nu7/2^+[624]$. The log ft values of the $\pi5/2^+[642] \rightarrow \nu5/2^+[633]$ transitions observed around the Am nuclei are 5.2 and 5.3 in the EC decay of ^{232}Np and ^{233}Np, respectively. The EC transition from ^{235}Pu to the $\pi5/2^+[642]$ ground state in ^{235}Np also shows a small log ft value of 5.4, which leads to the $\nu5/2^+[633]$ assignment to the ground state of ^{235}Pu. On the other hand, the EC transition from ^{235}Am to the $\nu5/2^+[633]$ ground state in ^{235}Pu shows log $ft > 6.0$, which implies that the ground state of ^{235}Am is not the $\pi5/2^+[642]$ state but the $\pi5/2^-[523]$. There are other experimental data for the $\pi5/2^+[642] \leftrightarrow \nu5/2^+[633]$ transition in the EC and β^- decays of ^{229}Pa and ^{231}Th with relatively large log ft values of 5.7 and 5.9, respectively. However, the present "lower limit" of log $ft > 6.0$ was derived from the $E1$ assumption for all the γ transitions except the 183.7 keV one. If some of other intense γ transitions are assumed as $M1$, which is considered more reasonable as suggested before and discussed later, the deduced ft value becomes about an order of magnitude larger. Therefore, the $\pi5/2^+[642]$ assignment is improbable for the ground state of ^{235}Am.

The level energy in ^{231}Np populated by the α-decay of ^{235}Am is indispensable to determine the atomic mass of ^{235}Am from the measured α-particle energy. Since the 6457 keV α transition is a favored transition with a hindrance factor of 1.2 [8], this transition populates the $\pi5/2^-[523]$ state in ^{231}Np. The 93rd proton of the ground state of $^{233-239}$Np occupies the $\pi5/2^+[642]$ orbital, and the $\pi5/2^-[523]$ state is located at 49.1, 59.5, and 74.7 keV in ^{235,237,239}Np, respectively [12]. Other orbitals lie at higher excitation energy. The spin-parity of the ground state of ^{231}Np is evaluated as $(5/2^\pm)$ [12]. Thus, it is highly probable that the ground state of ^{231}Np is either the $\pi5/2^+[642]$ state or the $\pi5/2^-[523]$. If the $\pi5/2^-[523]$ state is the ground state, the 6457 keV α transition becomes the ground-state–to–ground-state transition. On the other hand, if the $\pi5/2^+[642]$ state is the ground state, the $E1$ transition from the $\pi5/2^-[523]$ excited state to the $\pi5/2^+[642]$ ground state should be observed in coincidence with the 6457 keV α-particles. However, no α-coincident γ-ray peak was observed in the present experiment. By taking into account the detection efficiency for low-energy γ-rays and internal conversion coefficients of $E1$ transitions [11], the energy of the $\pi5/2^-[523]$ state in ^{231}Np is evaluated to be < 15 keV within a 99% confidence level. This limit is consistent with the fact that not only γ-rays but also L X-rays were not observed in coincidence with the α-particles, because the binding energy of L_3 electrons in the Np atom is 17.6 keV. Taking into account the α energy of 6457(12) keV, the level energy of < 15 keV, and an energy shift of the α peak centroid arising from the coincidence summing effect between the α-particle and following low-energy electrons, we have determined the Q_α value of ^{235}Am as 6569^{+19}_{-12} keV. Möller *et al.* [18] and Koura *et al.* [16] theoretically calculated the Q_α value of ^{235}Am as 6600 keV and 7237 keV, respectively. The measured value is in good agreement with the value calculated by Möller *et al.*

Spin-parities of the excited states in ^{235}Pu cannot be determined explicitly from the present experimental results. However, the γ-ray branching ratios compared with those of other $N = 141$ isotones ^{229}Ra, ^{231}Th, and ^{233}U [12,19] allow us to suggest tentative spin-parity and Nilsson orbital assignments. Figure 4 shows level energies of Nilsson single-particle states in $N = 141$ isotones. The ground states of all these isotones are known to be the $\nu5/2^+[633]$ state. The 41.9 keV level in ^{235}Pu is considered as the $7/2^+$ state in the $\nu5/2^+[633]$ band owing to its energy similar to those of the isotones. The 265.3 and 290.6 keV levels decay to both the $5/2^+$ and $7/2^+$ states in the $\nu5/2^+[633]$ band and not to the $9/2^+$ state. This fact suggests the $5/2^\pm$ assignment for these levels, because the low-lying $7/2^\pm$ states in ^{229}Ra, ^{231}Th, and ^{233}U decay to the $9/2^+$ state as well as the $5/2^+$ and $7/2^+$ states with substantial intensity, and the low-lying

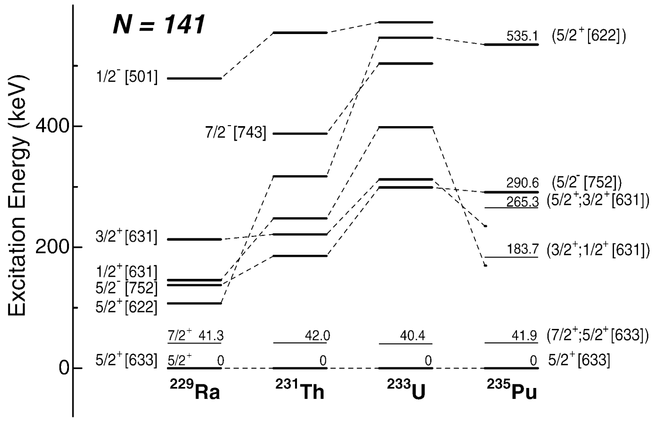

Fig. 4. Level energies of Nilsson single-particle states in $N = 141$ isotones.

$3/2^+$ states below 300 keV are not expected to decay to the $7/2^+$ state with observable intensity like those in the isotones. Thus, the $5/2^\pm$ states in the $\nu 5/2^-[752]$, $\nu 3/2^+[631]$, and $\nu 1/2^+[631]$ bands are the candidates for the 265.3 and 290.6 keV levels. On the other hand, the 183.7 keV level is considered as the $1/2^+$ or the $3/2^+$ state in the $\nu 1/2^+[631]$ or the $\nu 3/2^+[631]$ band because it decays only to the $5/2^+$ ground state. Moreover, the 183.7 keV level should belong to a band different from those of the 265.3 and 290.6 keV levels because the energy spacings between these levels are not suited for those of the $\nu 1/2^+[631]$ and the $\nu 3/2^+[631]$ bands.

The γ-ray branching ratios depopulating the $\nu 5/2^-[752]$ state to the $\nu 5/2^+[633]$ band in ^{229}Ra, ^{231}Th, and ^{233}U are $B(E1; 5/2^- \to 7/2^+)/B(E1; 5/2^- \to 5/2^+) = 0.30$, 0.41, and 0.34, respectively, which are in good agreement with 0.31(13) for the 290.6 keV level in ^{235}Pu. On the other hand, the ratios for the $5/2^+$ state in the $\nu 3/2^+[631]$ band in ^{231}Th and ^{233}U are $B(M1; 5/2^+ \to 7/2^+)/B(M1; 5/2^+ \to 5/2^+) = 1.15$ and 2.17, respectively. The branching ratio of 2.0(6) for the 265.3 keV level in ^{235}Pu is consistent with these values. The Alaga rule intensity ratio [20] for the $\nu 5/2^-[752] \to \nu 5/2^+[633]$ transition is $B(E1; 5/2^- \to 7/2^+)/B(E1; 5/2^- \to 5/2^+) = 0.40$, while that for the $\nu 3/2^+[631] \to \nu 5/2^+[633]$ is $B(M1; 5/2^+ \to 7/2^+)/B(M1; 5/2^+ \to 5/2^+) = 2.50$. These values also support the $\nu 5/2^-[752]$ assignment for the 290.6 keV level and the $\nu 3/2^+[631]$ assignment for the 265.3 keV level. The remaining $\nu 1/2^+[631]$ band should be assigned to the 183.7 keV level. Note that the energy of the 169.6 keV γ-ray which could not be placed in the decay scheme is consistent with that of the $1/2^+$ state in the $\nu 1/2^+[631]$ band if the 183.7 keV level is the $3/2^+$ state in the same band. Thus, we tentatively assign this $3/2^+$ state to the 183.7 keV level.

The EC decay of $^{237-240}$Am is characterized by the intense $\pi 5/2^-[523] \to \nu 5/2^+[622]$ transitions with $\log ft = 6.0$–6.2 [1–4]. This situation is also expected in ^{235}Am, that is, the EC transition to the $\nu 5/2^+[622]$ state in ^{235}Pu has the largest intensity with $\log ft \sim 6.0$. In order to estimate the $\log ft$ values to each of the levels, EC transition intensities are calculated using the following assumptions: i) all the γ transitions have the same multipolarity of $E1$, $M1$, or $E2$; ii) the multipolarities of all the γ transitions are $M1$ except for the 244.6, 248.6, and 290.6 keV $E1$ transitions. The latter assumption is based on the above spin-parity assignments. Under all the assumptions, the intensity of the EC transition to the 535.1 keV level becomes the largest with $\log ft = 5.7$–6.0. Thus, we assign the $\nu 5/2^+[622]$ configuration to the 535.1 keV level.

The proposed Nilsson orbital assignments are summarized in fig. 4. It was found that the energy spacings between the $\nu 5/2^+[633]$ ground state and other low-lying Nilsson states increase from ^{229}Ra to ^{233}U, then turn to decrease at ^{235}Pu. The increase would correspond to an increasing deformation from ^{229}Ra to ^{233}U, while the decrease at ^{235}Pu may suggest the opposite trend. A similar trend is found in the neighboring even-even $N = 142$ isotones. Energies of the first 2^+ states in the $N = 142$ isotones ^{230}Ra, ^{232}Th, ^{234}U, and ^{236}Pu are 57.4, 49.4, 43.5, and 44.6 keV, respectively [12], exhibiting the lowest energy at ^{234}U. These trends would imply the existence of a local deformation maximum around ^{234}U.

5 Conclusions

EC and α decays of ^{235}Am have been studied using the online isotope separator. Excited states in ^{235}Pu have been established for the first time, and tentative spin-parity and Nilsson orbital assignments were given to these levels. A local deformation maximum around ^{234}U has been suggested through the trends of excitation energies in the $N = 141$ and 142 isotones. The ground state of ^{235}Am was evaluated to be the $\pi 5/2^-[523]$ state from the deduced $\log ft$ value of the EC transition to the $\nu 5/2^+[633]$ ground state in ^{235}Pu. It was found that the $\pi 5/2^-[523]$ state in ^{231}Np is located at < 15 keV, which allowed us to determine the Q_α value of ^{235}Am as 6569^{+19}_{-12} keV.

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