^{233}U levels populated in the α decay of ^{237}Pu

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The 45.3-day nuclide ^{237}Pu decays primarily via electron capture. The only information available up to now concerning the nuclide's α-decay properties has been the observation of two α groups with energies of 5.65 and 5.36 MeV. In the present study, the photon spectrum of ^{237}Pu was investigated with x- and γ-ray Ge(Li) detectors. A total of 21 transitions was found to belong to ^{237}Pu decay; of these, 16 were assigned to its α-decay branch. An α-decay scheme was constructed on the bases of transition energy fits and known ^{233}U levels (including some observed only in reaction studies). The amounts of direct α-decay feedings were then calculated from intensity balances, and a total α-branching of $(4.2 \pm 0.4) \times 10^{-3}\%$ was deduced. α-decay hindrance factors identify the band heads of the (5/2)[752] and (7/2)[743] orbitals in ^{233}U at 298.9 and 503.5 keV, respectively. Contrastingly, the ^{233}Pa and ^{233}Np decay studies had assigned the (7/2)[743] state to the level at 298.9 keV.

[RADIOACTIVITY ^{237}Pu, measured E_γ, I_γ; deduced α/electron-capture ratio. ^{233}U, deduced levels, J^π.]

I. INTRODUCTION

Levels in ^{233}U have been investigated by means of inelastic deuteron scattering,[1,2] the ^{235}U(p,t) reaction,[3] the ^{234}U(d,t) and ^{234}U$(^3$He$,\alpha)$ reactions,[4] and the decays of ^{233}Np (Ref. 5) and ^{233}Pa (see, e.g., Ref. 6). Structure information from the α decay of ^{237}Pu, on the other hand, is sparse. Two α groups, 5.65 and 5.36 MeV, have been reported.[7] The first of these feeds the ground-state band, while the second group populates a level, or levels, at ~300 keV of excitation in ^{233}U.

The purpose of the present study was to obtain more information about the α-decay scheme of ^{237}Pu. This was done by investigating the nuclide's photon spectrum with the use of x- and γ-ray Ge(Li) detectors.

II. RESULTS

A 50-μm thick foil of ^{235}U (93% enrichment) was bombarded with ^{4}He ions accelerated in the Oak Ridge isochronous cyclotron. The incident energy on target was selected to be ~30 MeV so as to maximize the cross section for ^{237}Pu, the $(\alpha, 2n)$ product. Following irradiation, the plutonium fraction was chemically separated from the target material, fission products, and neptunium isotopes.

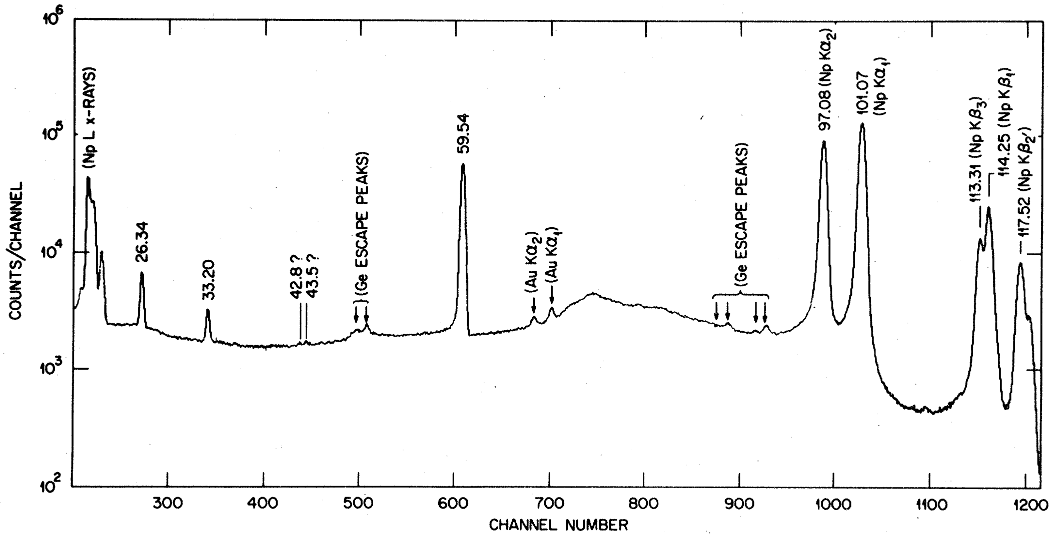

FIG. 1. Low-energy portion of the ^{237}Pu photon spectrum taken with an x-ray Ge(Li) detector. The 26.34-, 33.20-, 42.8-, 43.5-, and 59.54-keV γ rays are known to be transitions in ^{237}Np and are therefore assigned to ^{237}Pu electron-capture decay.

From an α-particle assay, it was determined that besides ^{237}Pu, the source contained ^{236,238,239}Pu. Because of their longer half-lives, the specific activities of ^{236,238,239}Pu were much less than that of ^{237}Pu. As a result, γ rays known to follow the decays of these three isotopes were not observed in our photon spectra.

Several singles γ-ray measurements, each about 60 h in duration, were made of this plutonium source over a period of three months. Background determinations were also made. γ rays were assigned to ^{237}Pu on the basis of the isotope's characteristic 45.3-day half-life. The photon energy region below ~120 keV was studied with an x-ray Ge(Li) detector 10 mm in diameter and 5 mm in depth with a resolution of 230 and 566 eV at 5.9 and 122 keV, respectively. A 54-cm^3 Ge(Li) detector with a resolution of 1.7 keV at 228 keV was used to obtain spectra of γ rays with energies above 100 keV. Portions of the x- and γ-ray spectra are shown in Figs. 1 and 2, respectively; peaks labeled by energy are assigned to ^{237}Pu. Transitions in ^{237}Np are well established, not only from study of ^{237}Pu electron-capture decay,[8] but also from ^{241}Am α decay (e.g., Refs. 9–11), ^{237}U β^- decay,[12] and ^{237}Np Coulomb excitation.[13] Five of these ^{237}Np transitions can be seen in Fig. 1. All of the

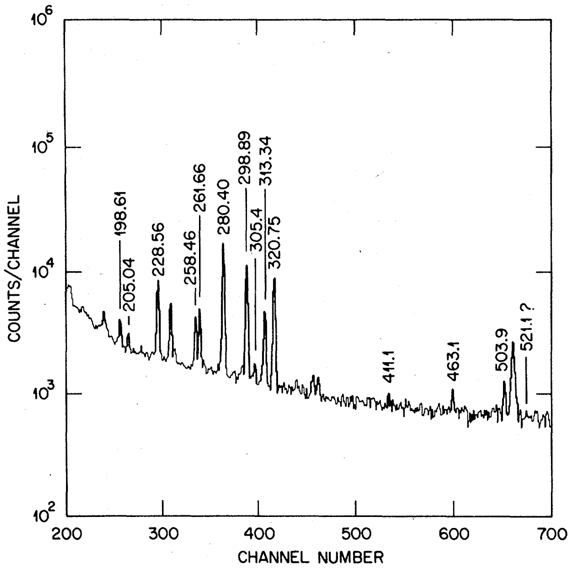

FIG. 2. Portion of the ^{237}Pu photon spectrum taken with a large-volume Ge(Li) detector. Peaks labeled by energy are assigned to the α decay of ^{237}Pu. Unlabeled peaks are due to background radiations.

TABLE I. Energies, intensities, and multipolarities of ^{237}Np transitions observed in the electron-capture decay of ^{237}Pu.

E_γ Previous work (Ref. 8)	E_γ Present work	I_γ[a] Present work	I_{ce} (selected lines only) Measured (Ref. 8)	Calculated	Multipolarity[b]	Total conversion coefficient[b]
	26.34 (5)	6.75 (20)			$E1$	9.0 (6) Anomalously converted
33.2	33.20 (5)	2.27 (7)	54 M_{12}	30 M_{12}	$M1+1.8\%\ E2$	188 (10)
	42.8 (1)	~0.090		~5.3 L	$(M1+E2)$[c]	~80[c]
43.5	43.5 (1)	~0.12	Very weak L_1, L_2	~15 L	$M1+14\%\ E2$	167 (12)
55.5	(55.52)[d]	<0.05	Very weak L_1, L_2		$M1+17\%\ E2$	68 (6)
59.6	59.54 (5)	100 (3)	13.1 L_3	13.1 L_3	$E1$	1.15 (7) Anomalously converted
76.4	(75.8)[d]	~0.0096[e]	<2 M	0.1 M	$(E2)$	54.5
	L x rays	1210 (40)				
	K x rays	1310 (40)				

[a] Relative intensities based on a value of 100 for the 59.54-keV transition. For absolute intensities per 100 ^{237}Pu decays, relative intensities should be multiplied by a factor of 0.0328 ± 0.0015.

[b] Multipolarities are based on the conversion-electron data of Refs. 9 and 12. Conversion coefficients are theoretical values from Hager and Seltzer and from Dragoun *et al.* (Ref. 16) except for the anomalously converted $E1$ 26.34- and 59.54-keV transitions. In these two instances experimental conversion coefficients are given.

[c] Conversion-electron data[9] from ^{241}Am α-decay for the 42.8-keV transition suggest $M1+\sim42\%\ E2$ with $\alpha \approx 400$. This large $E2$ admixture is not consistent with the fact that the electron lines for the 42.8-keV transition were not seen in ^{237}Pu decay (Ref. 8). If the $E2$ admixture were correct, the total L conversion-electron intensity would be twice that of the 43.5-keV transition. The indicated conversion coefficient was deduced from a ratio $E2/M1=0.0169$ calculated by using the strong coupling collective model and a ratio $E2/M1=0.018$ for the 33.20-keV transition.

[d] Not observed in the present study. Energies are from ^{241}Am α-decay (Ref. 11).

[e] Intensity calculated from the ratio $I_\gamma(42.8)/I_\gamma(75.8)$ as measured (Ref. 13) in the Coulomb excitation of ^{237}Np.

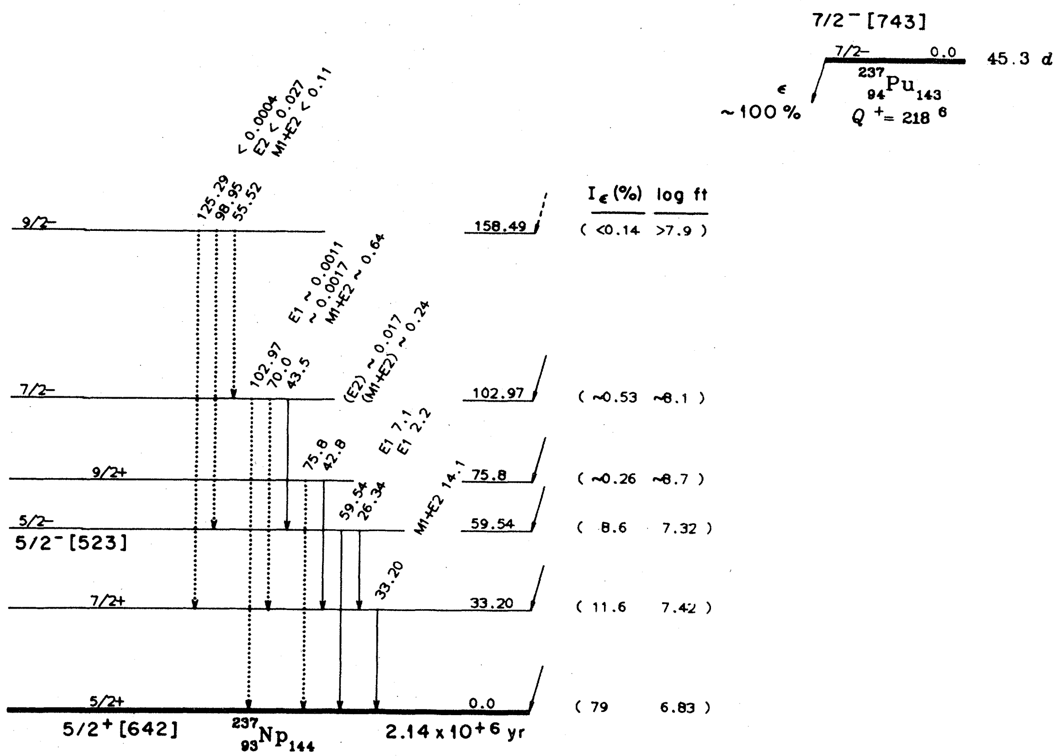

FIG. 3. Electron-capture decay scheme of ^{237}Pu. Numbers following energy values and multipolarities are total transition intensities calculated from photon intensities and internal conversion coefficients. These intensities are absolute values per 100 ^{237}Pu decays. Dotted lines indicate transitions not observed in the present study but are known from other decays (Refs. 9–12). Their intensities were calculated by using photon intensity ratios observed in ^{241}Am α decay (Ref. 11) and in Coulomb excitation (Ref. 13).

labeled γ-ray peaks in Fig. 2 are assigned to the α-decay of ^{237}Pu for the following reasons. Firstly, some of the transitions have been seen in ^{233}Np electron-capture decay[5] and/or ^{233}Pa β^- decay.[6] Secondly, the ^{237}Pu Q_{EC} is 218 ± 6 keV (Ref. 14) so that only levels below that energy in ^{237}Np can be populated. These levels, together with connecting transitions, are known from the investigations[8-13] mentioned above. The remainder of the peaks in Fig. 2 were also seen in the separate background measurements. They were found to be due to decay of ^{228}Th and ^{230}Th and their descendants.

Before discussing the ^{237}Pu α-decay scheme, the nuclide's predominant (>99.99%) electron-capture decay branch needs to be considered. This mode of decay has been investigated by Hoffman and Dropesky[8] and by Ahmad,[15] who studied primarily the electron spectrum. The electron data of Ref. 8 are summarized with our γ-ray results in Table I. Multipolarities are based on the conversion-electron measurements of Refs. 9 and 12. Conversion coefficients included in the table are theoretical values,[16] except for the anomalously converted E1 26.34- and 59.54-keV transitions. The electron-capture decay scheme of ^{237}Pu is shown in Fig. 3. Dotted lines in Fig. 3 indicate known transitions which were not observed in our study because of their weak intensities. Their strengths were calculated from the measured intensities of the 42.8-, 43.5-, and 55.5-keV γ rays and photon intensity ratios observed in ^{241}Am α decay[11] and in Coulomb excitation.[13] The relative electron-capture feedings to the excited states were then deduced from intensity balances. From these relative feedings, the experimental K x-ray intensity (Table I), and theoretical ratios of K-to-total capture, the amount of decay proceeding to the ^{237}Np ground state was determined. Note that in this particular decay, observed K x rays can only by due to K-capture because, with the exception of the weak 125-keV transition, all γ-ray energies are below the neptunium K electron binding energy. Finally, absolute intensities were obtained by normalizing the sum of all capture branches to 100%.

Table II lists the photon energies and intensities for ^{233}U transitions observed in the present study

TABLE II. Energies and photon intensities of ^{233}U transitions observed in the present study of ^{237}Pu α decay.

E_γ	I_γ [a]	Total conversion coefficient [b]
181.8 (10) [c]	~0.8 [c]	4.31 ($M1$)
198.61 (20)	7.3 (10)	0.1011 ($E1$)
205.05 (20)	3.2 (8)	3.08 ($M1$)
228.56 (20)	36.2 (15)	0.0730 ($E1$)
241 (2) [c]	~0.5 [c]	1.96 ($M1$)
258.46 (20)	16.1 (12)	0.0551 ($E1$)
261.66 (20)	18.1 (11)	0.0536 ($E1$)
280.40 (20)	100 (2)	0.0459 ($E1$)
298.89 (20)	72.2 (18)	0.0398 ($E1$)
305.4 (2)	2.9 (9)	0.0380 ($E1$)
313.34 (20)	27.8 (14)	0.0359 ($E1$)
320.75 (20)	59.6 (18)	0.0341 ($E1$)
411.1 (2)	1.7 (5)	0.0202 ($E1$)
463.1 (2)	3.4 (10)	0.0158 ($E1$)
503.9 (2)	6.9 (13)	0.0133 ($E1$)
521.1 (20)	~0.8	0.0125 ($E1$)

[a] Relative intensities based on a value of 100 for the 280.40-keV transition. For absolute intensities per 100 α decays, relative intensities should be multiplied by a factor of 0.218 ± 0.017.

[b] Theoretical conversion coefficients (Ref. 16); multipolarities were deduced from the proposed ^{237}Pu α decay scheme (Fig. 4).

[c] γ ray obscured by the presence of neighboring background radiation.

of ^{237}Pu α decay. On the bases of transition energy fits with levels known from reaction studies[1-4] and ^{233}Np and ^{233}Pa decay investigations,[5,6] a decay scheme was constructed (see Fig. 4). The relative photon intensities (Table II) were converted to absolute values per 100 ^{237}Pu decays (see discussion in the previous paragraph). Total transition intensities were calculated by using theoretical conversion coefficients.[16] α feedings to the levels at 298.89 keV and above were then deduced from intensity balances. Two a groups, 5.65 and 5.36 MeV with relative intensities of 21 ± 4 and 79 ± 8, respectively, observed by Thomas *et al.*[7] are presumed to feed the ground-state band and levels around 300 keV. These two relative intensities and the deduced feedings to the levels at 298.9, 320.8, and 353.7 keV were next used to calculate the amount of direct a feeding proceeding to the ground-state band. The sum of all the deduced feedings then yielded an α-decay branching ratio of $(4.2 \pm 0.4) \times 10^{-3}\%$ for ^{237}Pu. The previously reported value of $(3.3 \pm 0.3) \times 10^{-3}\%$ was calculated by Thomas *et al.*[7] from K x-ray intensities and the α activities of ^{237}Pu and ^{236}Pu in a radioactive source whose isotopic composition was known.

In Fig. 4 the intensities are expressed in terms of 100 ^{237}Pu α decays. The highly converted, low-energy, expected intraband transitions, not seen in our γ-ray spectra, are shown dotted in Fig. 4. Only three of them, 40.35, 51.5, and 92.0 keV, have actually been observed[6,13] experimentally. Because their intensities cannot be estimated realistically, feedings to individual band members indicated in Fig. 4 are not exact; total decay to each band, however, should be accurate.

III. DISCUSSION

The α-hindrance factors shown in Fig. 4 were obtained by using the spin-independent equations of Preston.[17] This formalism has proved to be useful for the understanding of α-decay systematics in the heavy-element region.[18] In fact, hindrance factors calculated with this theory have been adopted widely as criteria to be considered in spin assignments. In this formalism, hindrance factors depend on the nuclear radius parameter, r_0. The radii for even-even nuclei are determined by assuming the ground-state-to-ground-state α decays (s-wave transitions) to be unhindered. For all other transitions, hindrance factors are then computed by choosing their r_0 parameters to be the average of those for neighboring even-even nuclei. In the present case an r_0 of 1.5081 was used. It represents the average of the parameters for ^{232}U (1.5085) and ^{234}U (1.5078) as calculated in a recent survey[19] of α-decay systematics for $A \geqslant 204$.

The ground-state band in ^{233}U is well established[1,2,4-6] to be based on the $\frac{5}{2}$[633] Nilsson orbital. In decay studies[5,6] and Coulomb excitation[13] only the first three band members were observed. The $\frac{11}{2}^+$ state was seen in (d,d') scattering[1,2] at 153 ± 2 keV and in (d,t) and $(^3\mathrm{He},\alpha)$ reactions[4] at 156 ± 2 keV. Our γ-ray energies place the level at 155.1 ± 0.3 keV.

As mentioned in the previous section, due to the unobserved low-energy transitions, it is not possible to deduce α feedings to the individual members of the band. Hindrance factors for transitions connecting given orbitals are expected to exhibit similar patterns.[18] The ^{237}Pu and ^{235}U ground state orbitals are both $\frac{7}{2}$[743]. In the decay of ^{235}U to the ^{231}Th ground-state band, the hindrance factors[20] are about 2200, 1300, 1350, and 980 for the $\frac{5}{2}^+$, $\frac{7}{2}^+$, $\frac{9}{2}^+$, and $\frac{11}{2}^+$ states, respectively. By assuming the same pattern for ^{237}Pu decay and a total feeding of 20% to the ground-state band, the branchings to these states in ^{233}U are expected to be ~7.1, 7.2, 3.6, and 2.1, with hindrance factors of 2300, 1370, 1420, and 1880, respectively.

The (503.5 ± 0.4)- and (561.5 ± 1.0)-keV levels were observed in inelastic deuteron scattering[1]

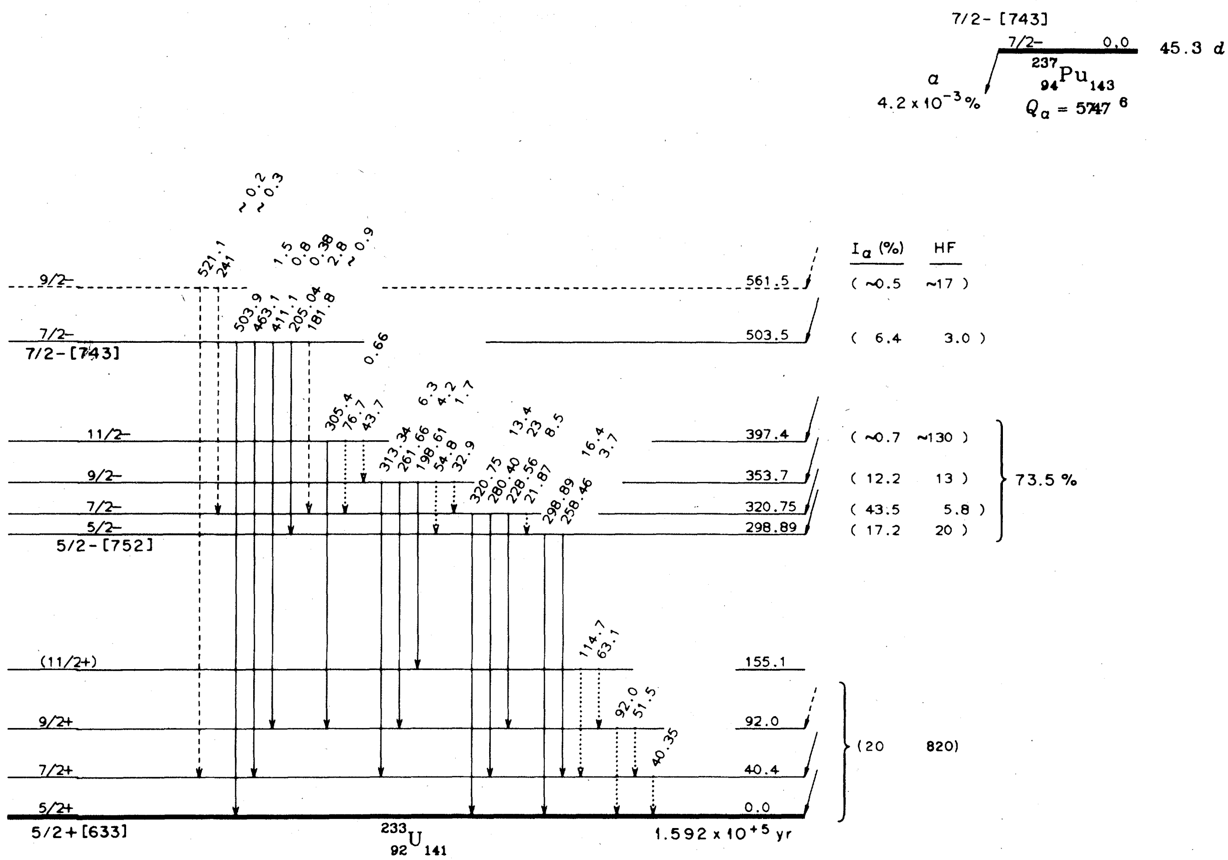

FIG. 4. α-decay scheme of ^{237}Pu. Numbers following energy values represent total transition intensities calculated from photon intensities and internal conversion coefficients. They are expressed in terms of absolute values per 100 ^{237}Pu α decays. Dashed lines represent uncertain transitions; dotted lines indicate transitions are expected but not observed in the present work. See text for a discussion of the α-decay branches to the various ^{233}U levels.

at 500 ± 2 and 560 ± 4 keV and in the ^{235}U(p, t) reaction[3] at 502.0 ± 1.5 and 567 ± 2 keV. The α decay to the 503.5-keV state is unhindered. Therefore, its orbital assignment is $\frac{7}{2}$[743], i.e., the same as that of its parent. The hindrance factor for the 561.5-keV level is consistent with its being the $\frac{9}{2}^-$ member. Our data thus confirm the assignments proposed in Refs. 1 and 3 for these two levels.

In the ^{233}Np and ^{233}Pa decay studies[5,6] the 298.9-keV level was proposed to be the $\frac{7}{2}$[743] state. However, this particular state is now established at 503.5 keV. Reaction data,[1,4] instead, have proposed the $\frac{5}{2}$[752] state at 298 ± 3 and 300 ± 3 keV. Our deduced hindrance factor for α decay to the (298.9 ± 0.2)-keV level confirms the $\frac{5}{2}$[752] state assignment. Further, we calculated the Nilsson hindrance factor for the 41.7-keV γ ray seen in ^{233}Pa decay to deexcite the 52-ps (Ref. 21) $\frac{5}{2}^+$, $\frac{3}{2}$[631] state at 340.2 keV. In the calculation, the $\frac{5}{2}$[752] configuration was used for the 298.9-keV level which is populated by the 41.7-keV transition. The low hindrance factor of 4.2 supports the adopted Nilsson assignment. In the decay of ^{233}Np, the 205-keV transition was placed[5] between this level and the $\frac{9}{2}^+$ member of the ground-state band. Given the correct spins and parities, the placement is improbable, and in Fig. 4 this transition, instead, is indicated to deexcite the $\frac{7}{2}^-$, 503.5-keV level.

The identification of the level at 320.75 keV as the $\frac{7}{2}^-$ member of the $\frac{5}{2}$[752] band agrees with the suggestion of previous investigators.[3-5] The $\frac{9}{2}^-$ and $\frac{11}{2}^-$ members were observed in the (d, d') reaction[1] at 353 ± 2 and 403 ± 4 keV and in the ^{235}U(p, t) reaction[3] at 352 ± 2 keV and ~396 keV. While the $\frac{9}{2}^-$ member was not seen in the (d, t) and (^{3}He, α) reactions,[4] the $\frac{11}{2}^-$ member was located at 398 ± 2 keV. Excitation energies determined in the present study for these states are 353.7 ± 0.2 and 397.4 ± 0.6

keV.

Strong Coriolis coupling is expected between the $\frac{5}{2}$[752] and $\frac{7}{2}$[743] bands. We calculated this Coriolis interaction from the observed level energies. The resultant mixing was found to be about 20%, in agreement with a mixing of ~25% estimated by Friedman *et al.*[3] from their (p,t) reaction data. Strong couplings with $\frac{3}{2}$[761] and $\frac{9}{2}$[734] bands are also expected. However, these states have not as yet been observed in ^{233}U.

To conclude, we would like to point out that the current mass evaluation of Wapstra and Bos[14] assumes that the 5.65-MeV ^{237}Pu α particle proceeds to the ^{233}U ground state. Our investigation suggests that the first-excited state is populated about as intensely as the ground state. A more accurate determination of the ^{237}Pu α-particle energies should resolve this point. An investigation with the use of a magnetic α spectrometer would thus be desirable.

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