Decay of 37.5-min ^{236}Th and 9.1-min ^{236}Pa

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The decays of ^{236}Th and its ^{236}Pa daughter were studied by γ-ray spectroscopy. By means of radiochemical methods and a continuous separation technique pure sources of parent and daughter were prepared so that each was free (or almost free) of the other. For ^{236}Th, 14 of the 17 observed γ rays are new; for ^{236}Pa 16 new γ rays were found. Absolute intensities of all the γ rays were determined with the aid of 4π beta measurements on ^{236}Pa. A new decay scheme is proposed for ^{236}Th and a considerably expanded one is given for ^{236}Pa. The half-lives determined are 37.5±0.2 min for ^{236}Th and 9.1±0.1 min for ^{236}Pa.

I. INTRODUCTION

The identification of 37-min ^{236}Th was first reported in 1973 independently by Orth *et al.*[1] and by Kaffrell and Trautmann.[2] The former produced ^{236}Th by the (p,3p) reaction on ^{238}U with 100-MeV protons, while the latter produced it with 140-MeV bremsstrahlung on ^{238}U through the (γ,2p) reaction. They chemically separated the thorium and observed the growth decay of the previously studied 9-min ^{236}Pa daughter activity.[3,4] Orth *et al.*[1] also performed electromagnetic isotope separation on their thorium samples in order to remove strong interference from neutron-deficient thorium isotopes. They were able to assign four γ rays to the decay of ^{236}Th and they proposed a partial decay scheme.

In this paper we report on considerably more extensive studies of the γ-ray spectra and decay properties of both ^{236}Th and ^{236}Pa. Neutrons (30—160 MeV) from the Brookhaven medium energy intense neutron facility (MEIN),[5] were used to produce ^{236}Th by the ^{238}U(n,2pn) reaction while production of interfering neutron-deficient

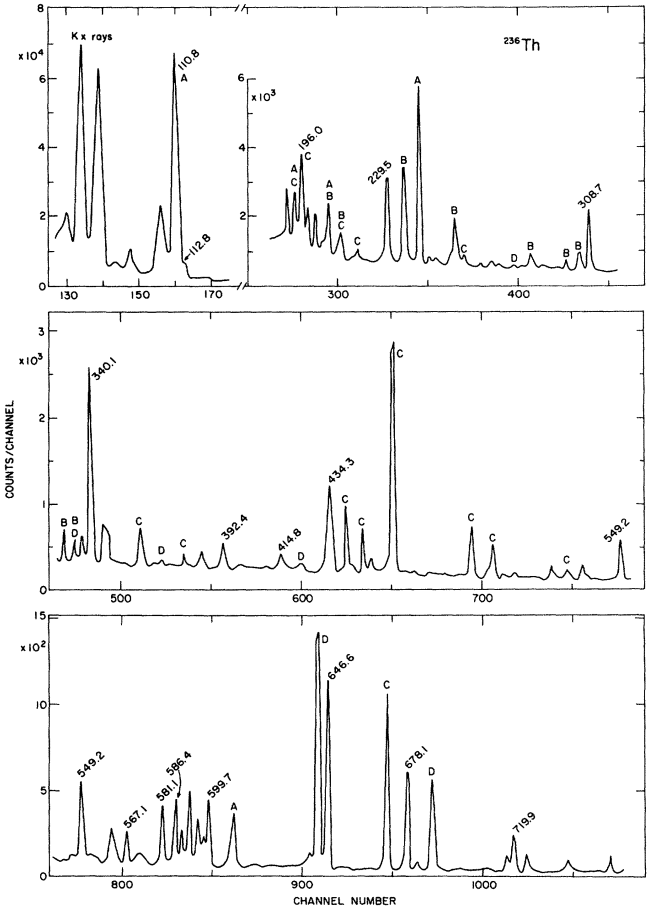

FIG. 1. Spectrum showing γ rays from the decay of 37.5-min ^{236}Th; energies are shown in keV. Peaks from other nuclides are identified by the following letters: *A*, 31-min ^{226}Th; *B*, 18.7-d ^{227}Th; *C*, 22-min ^{233}Th; *D*, 9.1-min ^{236}Pa.

TABLE I. Gamma-ray energies E_γ and intensities I_γ following decay of 37.5-min ^{236}Th. I_γ as γ rays per 100 disintegrations.

E_γ (keV)	I_γ This work	I_γ Ref. 4
110.8(1)	4.2(12)	2.9(8)
112.8(2)	0.24(9)	0.7(3)
131.6(10)[a]		0.56(28)
196.0(1)	0.69(14)	
229.5(1)	0.56(8)	0.56(28)[b]
308.7(1)	0.42(5)	
340.1(1)	0.67(9)	
392.4(1)[a]	0.17(3)	
414.8(3)[a]	0.13(3)	
434.3(1)[a]	0.67(9)[c]	
549.2(1)	0.32(9)	
567.1(3)	0.13(3)	
581.1(2)	0.20(4)	
586.4(2)[a]	0.09(4)	
599.7(1)[a]	0.24(3)	
646.6(1)	0.72(11)	
678.1(1)	0.47(7)	
719.9(1)[a]	0.21(3)	

[a]Not placed in level scheme.
[b]Normalized to I_γ=0.56 found in this work.
[c]This γ ray, from a ^{236}Th-^{236}Pa equilibrium source, was used for normalizing the I_γ values (I_γ=37.0 for the 642.3-keV γ ray of 9.1-min ^{236}Pa).

Th isotopes was minimized. Chemical separations, including the continuous milking technique,[6] were used to greatly reduce interference from the 9-min ^{236}Pa daughter γ rays when studying the 37-min ^{236}Th parent, and likewise, to remove all Th activities when measuring the γ radiation of ^{236}Pa. In this way it was possible to uncover many more γ rays and improve the measurements of energies and intensities of previously observed transitions. Modified and expanded decay schemes are proposed and discussed.

II. EXPERIMENTAL

Targets of natural uranium metal (1—2 g) were irradiated 30—60 min with spallation neutrons (30—160 MeV, $\sim 1\times 10^{11}$ n/cm^2 sec) at the MEIN facility.[5] In one case a more intense source of ^{236}Th was prepared by irradiating 0.5g of U metal directly with the 200-MeV proton beam (5 μA) for 30 min (the p,3p reaction).

The most important steps of the chemical separation procedure for ^{236}Th include the following: dissolution of U in 12 N HCl, fuming to dryness with HNO_3+HClO_4, adsorption of Th^{+4} onto an MP-1 anion exchange column from 10*N* HNO_3 (while U, Np, and rare earths pass through), elution of Th^{+4} with 12N HCl, extraction of Th from the acetic acid buffer (pH 3) into the TTA reagent, and back extraction with 1.0*N* HCl. Chemical yields were usually $\sim$70% and were estimated from the ^{234}Th activity present in natural U targets. The γ-ray measurements started about 45 min after the end of bombardment. For

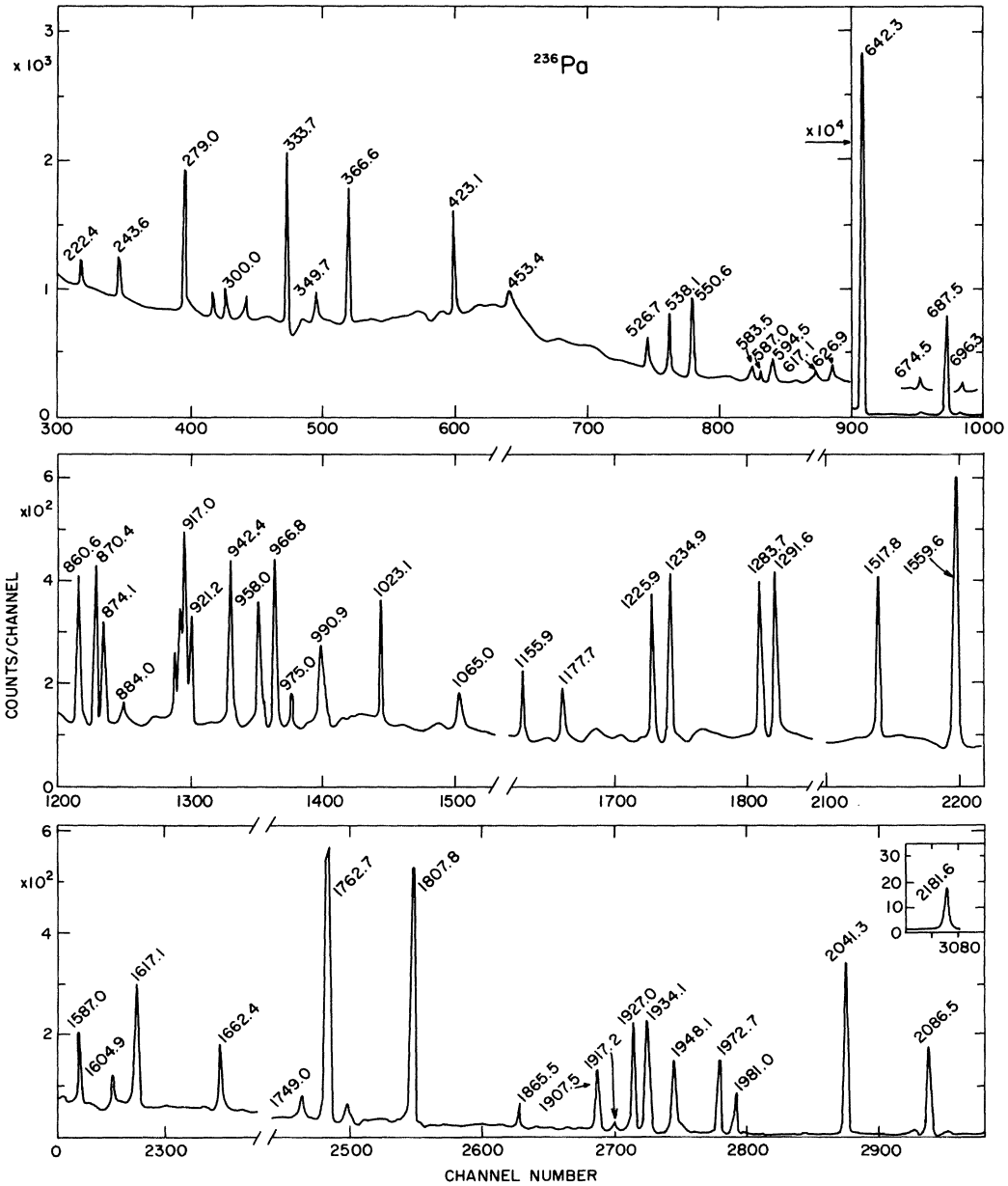

FIG. 2. Spectrum showing γ rays from the decay of 9.1-min ^{236}Pa; energies in keV.

the preparation of ^{236}Pa isolated from its parent the purified ^{236}Th was absorbed from 1.8N HCl onto a DOWEX-50 cation exchange column and the Pa daughter was eluted off with the same solution, which was then fed directly through a 2 cm^3 flat cell placed near a Ge(Li) detector. Alternatively, the solution was collected and the ^{236}Pa coprecipitated with $Fe(OH)_3$, or the solution was collected and evaporated on a very thin plastic film (10 μg/cm^2) to prepare a ^{236}Pa source for 4π beta counting. For obtaining relatively "clean" ^{236}Th γ-ray spectra, the DOWEX-50 cation exchange column itself was placed near the Ge(Li) detector while ~95% of the ^{236}Pa daughter activity was being flushed away.

The γ-ray spectra were measured with calibrated Ge(Li) detectors (50 cm^3, FWHM ~2.0 keV at 1332 keV). For detailed observation of the lower energy γ and x ray radiation (< 150 keV), a 1-cm thick Ge detector was used. The spectra were recorded on magnetic tapes with a computerized 4096 channel analyzer system and were analyzed later with the INTRAL code.[7] Decay curve analysis was done with the CLSQ code.[8] For measurements of total disintegration rates a 4π beta gas-flow proportional counter was used.

TABLE II. Gamma-ray energies E_γ and intensities I_γ following decay of 9.1-min ^{236}Pa. I_γ as γ rays per 100 disintegrations.

E_γ (keV)	I_γ This work	I_γ Ref. 4	E_γ (keV)	I_γ This work	I_γ Ref. 4
45.2			975.0(2)	0.19(5)	
68.8[b]	≤0.3	a	990.9(2)[b]	0.55(6)	a
104.3(1)	<0.15	a	1006.3(5)	<0.15	
222.4(1)	<0.21		1023.1(3)	0.58(5)	a
243.6(2)	0.23(3)	0.2	1065.0(2)	0.32(4)	0.30
279.0(1)	0.53(3)	0.51	1155.9(1)[b]	0.40(5)	a
300.0(1)	0.15(3)	0.07	1177.7(2)	0.36(5)	a
333.7(1)	0.82(4)		1225.9(1)	0.80(6)	a
349.7(2)[b]	0.23(4)		1234.9(1)	1.09(7)	a
366.6(1)	0.78(9)	0.6	1283.7(1)[b]	1.14(7)	a
423.1(1)	0.95(5)	0.63	1291.6(1)	1.09(7)	a
453.4(5)[b]	<0.5		1517.8(1)[b]	1.25(7)	a
526.7(2)	0.31(3)	a	1559.6(1)	2.2(2)	2.3
538.1(1)	0.58(9)	0.4	1587.0(2)[b]	0.66(6)	a
550.6(1)	1.08(6)	a	1604.9(2)	0.4(1)	
583.5(2)	<0.2		1617.1(1)	0.91(8)	1.07
587.0(2)	<0.2		1662.4(2)	0.60(6)	0.63
594.5(3)	0.32(5)	0.2	1749.0(2)[b]	0.33(4)	a
617.1(2)	0.21(4)		1762.7(1)	6.0(3)	6.7
626.9(2)[b]	0.23(5)		1773.5(3)[b]	0.30(3)	a
642.3(1)	37.0(20)	37[c]	1807.8(1)	2.24(12)	2.48
674.5(2)	0.21(7)		1865.5(2)	0.24(3)	a
687.5(1)	9.9(5)	9.5	1907.5(1)[b]	0.60(8)	a
696.3(2)[b]	0.19(4)		1917.2(2)[b]	0.06(2)	
740.8(2)[b]	0.33(5)	a	1927.0(2)	1.02(7)	a
860.6(1)	0.76(2)	a	1934.1(2)	1.07(7)	a
870.4(2)	0.69(6)	a	1948.1(2)[b]	0.91(9)	a
874.1(2)	0.51(5)	0.4	1972.7(1)	1.02(9)	a
884.0(2)	<0.15		1981.0(3)	0.51(5)	a
917.0(3)	1.37(8)	a	2041.3(1)	1.67(9)	1.9
921.2(2)	0.4(1)	0.31	2078.5(5)	<0.09	0.11
942.4(2)	0.87(6)	0.7	2086.5(2)	0.93(8)	1.00
958.0(2)	0.84(9)		2181.6(3)	0.18(5)	0.06
966.8(2)	0.91(8)	0.78			

[a] γ ray observed but intensity not reported.
[b] Not placed in level scheme.
[c] Normalized to I_γ = 37.0 at 642.3 keV.

III. RESULTS

In order to establish γ-ray intensities on an absolute basis (γ's per disintegration), it was necessary to prepare pure carrier-free sources of 9-min ^{236}Pa and measure both their absolute β-decay rates (4π counter) and their absolute γ-decay rates [642.2 keV with a calibrated Ge(Li) detector]. In four separate experiments, pairs of ^{236}Pa sources were prepared, a stronger one for the γ measurement and a weaker one for the 4π β measurement. Corrections of a few percent were made for the presence of a long-lived component in the decay of the β sources. The strong and weak sources were related to each other via the convenient 312-keV γ ray of 27-d ^{233}Pa. The weighted mean of the absolute intensity of the 642.3-keV γ ray of ^{236}Pa was found to be 37±2 per 100 disintegrations. The intensities of all the other γ rays were then normalized to this value. The γ-ray intensities of the parent 37-min ^{236}Th were also put on an absolute basis through measurements of sources in which parent and daughter were in equilibrium; proper account was taken of the parent/daughter ratio (0.76).

Figure 1 shows the γ spectrum of a purified thorium source from which the protactinium was being removed continuously. The γ rays belonging to the decay of ^{236}Th are indicated by the energy in keV labeled above the peaks. Each of these was shown to decay with the proper half-life. Peaks belonging to other nuclides are designated by the code A–D, which is explained in the caption. The ^{236}Pa daughter peaks (D) are suppressed by a factor of ~20 below the equilibrium values, and uranium K x-ray peaks are absent. Table I shows the ^{236}Th γ rays observed in this work and in the experiments of Orth *et al.*;[1] uncertainties in the last digits are given in parentheses, and the energies are those determined here; the I_γ values are all related to the absolute intensity of 37.0 for the 642.3-keV γ ray of the ^{236}Pa daughter. The intensity of the 110.8-keV γ ray was corrected for contributions from ^{226}Th (111.1 keV) and from Pa K_{β_2} x rays (111.5 keV). The 112.8-keV γ ray observed in this work decayed consistent with a 37-min half-life. Orth *et al.* observed the 131.6-keV γ ray in their spectrum of mass separated ^{236}Th, but in our Th sources this peak was masked by contributions from other isotopes. The 14 new γ rays reported here were revealed mainly because of the continuous milking technique which suppressed the γ radiation of the 9-min ^{236}Pa daughter activity. An accurate value of the ^{236}Th half-life was obtained by carefully following the decay of four ^{236}Th-^{236}Pa equilibrium sources. The weighted average for $T_{1/2}$ obtained from the two most intense peaks (642.3 and 687.5 keV) is 37.5±0.2 min, which is in good agreement with 37.5±1.5 and 36±3 min reported previously.[1,2]

Figure 2 shows the γ-ray spectrum of ^{236}Pa being continuously separated from its ^{236}Th parent. All γ rays attributed to ^{236}Pa are labeled with the energy in keV above

the peaks. Table II compares our measured absolute intensities I_γ with those reported previously[3,4] (renormalized to $I_\gamma=37.0$ for the 642.3-keV peak). The E_γ values are those determined here, and uncertainties in the last digits are shown in parentheses. Upper limits shown for I_γ indicate that the peaks were clearly observed but their attribution to ^{236}Pa is not certain. For those γ rays where Trautmann *et al.*,[3] reported intensities, agreement with our I_γ values is excellent. In addition, we found 16 new γ rays and we measured I_γ for 26 γ rays observed by Trautmann *et al.*, but for which they did not report intensities. The well-known 45.2-keV transition was not seen because it is highly converted. By carefully following the decay of the 642.3- and 687.5-keV γ rays in five highly purified ^{236}Pa sources, the half-life was determined to be 9.1 ± 0.1 min, in agreement with 9.1 ± 0.2 min reported previously.[3]

IV. DISCUSSION

Based on the ^{236}Pa decay data of Trautmann *et al.*,[3] Schmorak[4] proposed a tentative decay scheme. With this as a framework we added the new data and now propose the expanded and modified version shown in Fig. 3, into which about 75% of the γ transitions are accommodated. From a balance of the γ-transition intensities it follows that 48% of the ^{236}Pa β decay is to the 1^-, $K^\pi=0^-$, 3.78-nsec state in ^{236}U at 687.5 keV. Another 30% decay to higher levels, and about 20% decay to the 0^+ and 2^+ levels of the $K^\pi=0^+$ ground state band. The 56.5-keV transition between the 744.0- and the 687.5-keV levels was not observed here (because it is highly converted), but it was seen previously[4] in the decay of the 100-nsec ^{236}U isomer at 1053 keV.

Numerous levels in ^{236}U were discovered over many years[4] by means of various nuclear reaction studies[9-13] and radioactivity experiments.[3,4] Each process is usually selective in terms of the kind of excited states which are produced. Coulomb excitation, (p,p') and (d,d') reactions, and two-nucleon transfer reactions produce mostly collective states with $\pi=(-1)^I$ and $I=0^+,2^+,4^+,\ldots,1^-,3^-,5^-,\ldots,$. On the other hand, (d,p) reactions excite mostly two-particle negative parity neutron states which involve the unpaired neutron from the target ^{235}U ground state, $\frac{7}{2}^-[743]_n$. In (n,γ) reactions the neutron-capture states usually have $I^\pi=3^-$ or 4^-, and they depopulate by $E1$ transitions to $I^\pi=2^+-5^+$ states. Beta decay is also selective because the final states produced are mostly those with $I=I_0,I_0\pm1$ (where I_0 is the parent spin). Thus β decay of ^{236}Pa, with $I_0=1$, provides a very useful means of preferentially populating low-spin states in ^{236}U. In this way seven additional new levels were identified above 1110 keV.

Assignment of the ^{236}Pa ground state to $I_0=1$ and $K=0,1$ was based on the following considerations: 48% of all the β decays ($\log ft=6.8$) are to the $I=1^-$, $K=0$ level at 687.5 keV; an $I_0=2$, $K=2$ assignment is ruled out

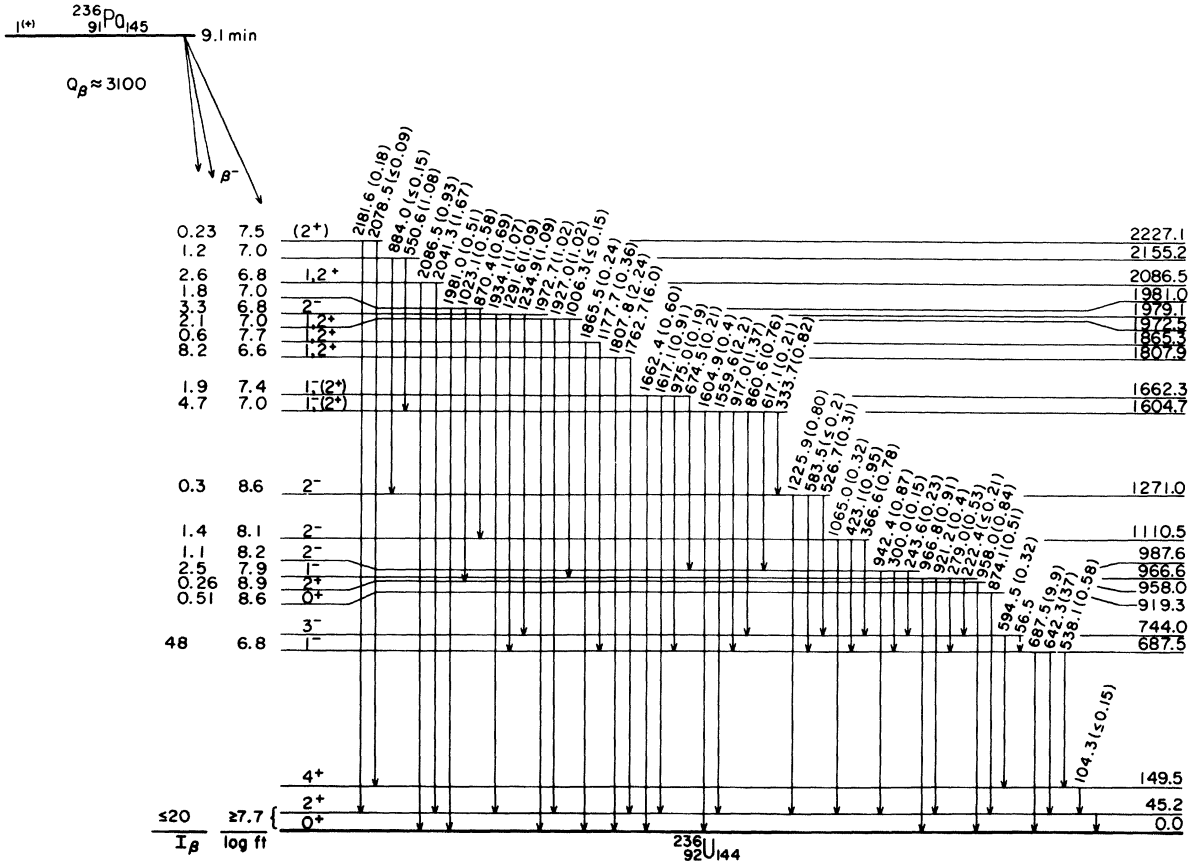

FIG. 3. Decay scheme proposed for the decay of 9.1-min ^{236}Pa; deexcitation of levels in ^{236}U.

because this β transition to the 687.5-keV level is not K forbidden; and an $I_0=0$, $K=0$ assignment is ruled out because $>6\%$ of the β decays are to $I=2$ levels. The probable positive parity assignment was deduced by taking into account the microscopic structure of the states connected by the β transitions. According to the Nilsson level diagram, the $K^\pi=0^-$ octupole state of ^{236}U may be formed mostly by the configuration closest to the Fermi surface: $\frac{7}{2}^-[743]_{n_1},\frac{7}{2}^+[624]_{n_2}$ and $\frac{5}{2}^+[622]_{n_1},\frac{5}{2}^-[752]_{n_2}$. However, weak excitation of this level in the ^{235}U(d,p) reaction, with a $\frac{7}{2}^-[743]_n$ ground state, suggests that the former component is weak and therefore that the latter component is the major one. The large I_β ($\log ft=6.8$) may therefore be related to this final configuration. The parent configuration for $^{236}_{91}Pa_{145}$ may be deduced in the usual way from the known Nilsson configurations of neighboring odd Z and odd N nuclei: $\frac{1}{2}^+[400]_p$ or $\frac{1}{2}^-[530]_p$, and $\frac{1}{2}^+[631]_n$. However, these configurations would result in a strongly retarded β transition to the $K=0^-$ octupole state, $\frac{5}{2}^+[622]_{n_1},\frac{5}{2}^-[752]_{n_2}$, because it would require simultaneous change of two particles. Also, β transition to the $K=0^+$ ground state would be unhindered first forbidden. Neither of these predictions is supported by the observations: $\log ft=6.8$ for the former transition and $\log ft\geq 7.7$ for the latter. The most probable ^{236}Pa ground state configuration is $\frac{3}{2}^+[651]_p,\frac{5}{2}^+[622]_n$. The β transition to the 1^- octupole state is then a one-particle unhindered first forbidden transition

$$\tfrac{3}{2}^+[651]_p,\tfrac{5}{2}^+[622]_n\rightarrow\tfrac{5}{2}^-[752]_{n_1},\tfrac{5}{2}^+[622]_{n_2},$$

with $\Delta\Omega=1$, $\Delta n_Z=0$, and $\Delta\Lambda=1$, and β transition to the $K=0$, $I^\pi=0^+$ ground state is the strongly hindered allowed transition $\frac{3}{2}^+[651]_p\rightarrow\frac{5}{2}^+[622]_n$. Thus, the spin and parity of the ^{236}Pa ground state can be assigned as $I^\pi=1^{(+)}$.

The lifetime of the $K=0^-$, $I^\pi=1^-$ state in ^{236}U is known to be unusually long[4] (3.78 nsec), with $B(E1;1^-\rightarrow 0^+)\approx 10^{-8}$ W.u. instead of 10^{-4}–10^{-5}, which is normal for actinides. The reasons for such strong retardation are not known, but it is interesting to examine the experimental and theoretical ratios $[B(E1;1^-\rightarrow 0^+)]/[B(E1;1^-\rightarrow 2^+)]$ for transitions from the $K=0^-$, $I=1^-$ and the $K=1^-$, $I=1^-$ octupole states to the $K=0$, $I=0^+$ and 2^+ states, respectively. For the 687.5-keV $K=0^-$, $I=1^-$ level the experimental ratio is 0.22, while the theoretical Alaga rule prediction[14] is 0.5; for the 966.6-keV $K=1^-$, $I=1^-$ level the experimental ratio is 1.97 and the theoretical one is 2.0. Thus for the 687.5-keV $K=0^-$ state in ^{236}U a large nonadiabatic effect influences both the absolute values of $B(E1)$ and their ratios.

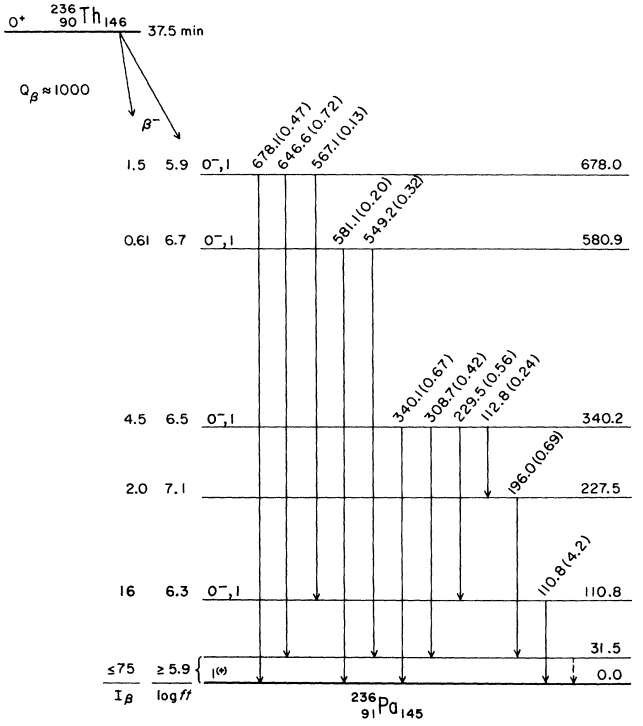

FIG. 4. Decay scheme proposed for the decay of 37.5-min ^{236}Th; deexcitation of levels in ^{236}Pa.

In Fig. 4 we propose a tentative decay scheme for ^{236}Th in which 11 of the 18 γ ray transitions are included. Placement of each level is supported by observation of two to four ingoing and/or outgoing γ rays. $Q_{\beta^-}\approx 1.0$ MeV was taken from the estimate in Nuclear Data Sheets,[4] and the β-ray branchings I_β were calculated with the assumption that the γ transitions are all $M1$. The relatively small values of $\log ft$ (≤ 6.7) for most of the observed β branches suggest that $I=1$ or 0^- for the corresponding levels in ^{236}Pa. Orth *et al.*[1] suggested a different partial decay scheme, but theirs was based on only four observed γ rays.

ACKNOWLEDGMENTS

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