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Decay of 7.3-min ^{235}Th and 24.6-min ^{235}Pa

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Sources of ^{235}Th and ^{235}Pa were prepared by medium energy neutron and proton irradiations of uranium followed by radiochemical separations. Gamma-ray spectroscopy showed 13 new γ rays in the decay of ^{235}Th. Its half-life is 7.3 ± 0.1 min, and that of ^{235}Pa is 24.6 ± 0.2 min. A partial decay scheme is proposed for ^{235}Th, and a large anomalous retardation is reported in one branch of the ^{235}Pa β^- decay.

I. INTRODUCTION

^{235}Th was first identified by Trautmann, Denig, and Herrmann[1] who produced it, via the ^{238}U(n,α) reaction, by irradiating a uranium target with 14.8-MeV neutrons. By means of time-sequenced chemical separations of the previously known 24-min ^{235}Pa daughter activity[2] the ^{235}Th half-life was determined to be 6.9 ± 0.2 min. In subsequent studies[3,4] these authors, in collaboration with Kaffrell, reported five γ rays associated with ^{235}Th decay and about ten γ rays following decay of ^{235}Pa. Relative intensities of the γ rays were not given. A recent compilation[5] summarizes these prior investigations.

In this paper a new study of these two nuclides is described.[6] A level scheme is proposed for $^{235}_{91}$Pa by combining the new γ ray data with previous (t, α) reaction data and with the known properties of lighter odd-A Pa nuclei. Although decay of $^{235}_{91}$Pa is to $^{235}_{92}$U whose level scheme is known in great detail,[5] a very large anomalous retardation was observed in one of the β^- transitions.

II. EXPERIMENTAL

Sources of ^{235}Th were prepared by irradiating 1—2 g of natural U metal for 10—15 min either with 30—160 MeV neutrons at the Brookhaven Medium Energy Intense Neutron facility[7] (MEIN) or with 14-MeV neutrons from the 60-inch BNL isochronous cyclotron. To prepare stronger sources of ^{235}Pa, its parent ^{235}Th was produced by irradiating 150 mg of U for 10 min with a 5 μA proton beam from the 200-MeV linear accelerator.

Thorium activity was separated from the irradiated uranium by adsorption on anion exchange columns and further purified by extractions with thenoyl trifluoroacetone (TTA) reagent.[6] Then Th in 6M HCl was extracted into 2—3 ml of HDEHP (bis-2-ethylhexylorthophosphoric acid) solution for the γ-ray measurements which started $\sim$28 min after the end of the bombardment. Sources of ^{235}Pa were prepared by separation from its ^{235}Th parent. The Th activity, partially purified by the anion exchange step, was loaded onto a cation exchange column and Pa activity was eluted by 1.8M HCl. This fraction was then purified further[1,4] by extracting with DIBC (diisobutylcarbinol) from 12M HCl and back-extracted with a mixture of 12M HCl and 0.5M HF. Finally Pa was coprecipitated with $Fe(OH)_3$. Chemical yields were about 60% for Th and $\sim$80% for Pa.

The γ-ray spectra were measured with calibrated Ge(Li) detectors ($\sim$50 cm^3, FWHM $\sim$2.0 keV at 1332 keV). The measurements were made with the samples close to the detector because of the limited source strength. Spectra were recorded on magnetic tape with multichannel analyzer and γ-ray energies and intensities were obtained later by means of the INTRAL code.[8] Decay curve analysis was performed with the least squares CLSQ code.[9] Beta-ray measurements were made with an end-window gas flow proportional counter.

III. RESULTS

The ^{235}Th sources prepared from the 30—160 MeV neutron irradiations were of reasonably good strength, but the γ-ray spectra showed some interference from other Th isotopes: 8-min ^{225}Th, 31-min ^{226}Th, 22-min ^{233}Th, and 37.5-min ^{236}Th. The ^{235}Th sources from 14-MeV neutron irradiations showed only a few extraneous peaks in their γ-ray spectra, but these sources were weak (Fig. 1). Seventeen γ rays were found to decay with the proper half-life and thus are attributed to the decay of ^{235}Th. The unlabeled peaks in Fig. 1 are from impurities; no peaks can be assigned to the ^{235}Pa daughter. Table I shows the γ-ray energies and intensities with their associated uncertainties in the last digits given in parentheses. The data are based on weighted averages from four runs, two each from the 14-MeV and the 30—160 MeV neutron irradiations. The γ-ray energies which had been reported by Trautmann, Denig, and Kaffrell[3] are 416.2, 659.4, 727.2, 747.0, and 931.8 keV. These are generally about 0.8 keV lower than those reported here (659.4 keV γ not seen at all). A *rough* value of the absolute intensity of the 417.0-keV γ rays is $\sim$2 per 100 disintegrations. This was calculated by comparing ^{235}Th γ-ray intensities with the known absolute intensities[6] of ^{236}Th-^{236}Pa and then estimating the relative cross sections (4/1) for producing ^{235}Th and ^{236}Th from ^{238}U bombarded by 30—160 MeV neutrons.[10] Absolute intensities for the ^{235}Th γ rays estimated in this way are probably reliable to within a factor of 2 or better. By carefully following the decay of the most intense γ ray (417.0 keV) in four sources, the ^{235}Th

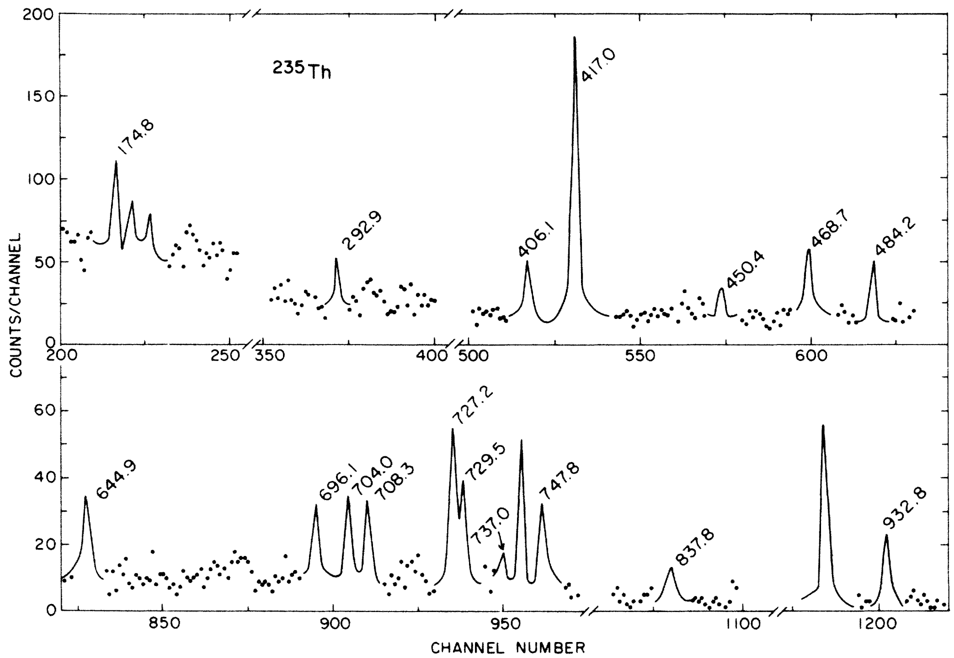

FIG. 1. Spectrum showing γ rays from decay of 7.3-min ^{235}Th; energies are in keV. The unlabeled peaks are from impurities.

half-life was determined to be 7.3±0.1 min. This compares with 6.9±0.2 min determined previously[2] from the milking experiments.

An accurate value of the ^{235}Pa half-life was determined by carefully following the β decay of six highly purified sources milked from ^{235}Th prepared by the ^{238}U(n,α) reaction with 14-MeV neutrons. The weighted average is 24.6±0.2 min, slightly longer than the adopted value[5] of 24.1±0.2 min. Kaffrell *et al.*[4] had reported several γ rays (127.8–659.3 keV) following decay of ^{235}Pa but in the present work no clear evidence was found for these even in the stronger Pa sources ($\geq 2\ \mu$Ci) separated from the Th parent produced in 200-MeV proton irradiated uranium. The two most intense γ rays reported previously,[4] at 375 and 414 keV, were just barely observed here, hardly above background. By comparison with the 312-keV γ ray of 27-d ^{233}Pa (daughter of 22-min ^{233}Th) upper limits for their absolute intensites were estimated to be below 0.01 percent. The previous limit[4] was $<3\%$. Thus, $>99.99\%$ of the ^{235}Pa decay must be to low-lying states in ^{235}U.

IV. DISCUSSION

The only previous data on the energy levels of ^{235}Pa were derived from the ^{236}U(t,α) study of Thompson *et al.*[11] By combining their results with the γ-ray data of Table I, and with consideration of nuclear systematics, a partial decay scheme was constructed (Fig. 2) which includes 11 of the 17 observed γ-ray transitions.

The ground state of ^{235}Th has been assigned[5] to $\frac{1}{2}^{+}$[631], consistent with the assignment in other odd-A, $N=145$ nuclei, i.e., $^{237}_{92}$U$_{145}$ and $^{239}_{94}$Pu$_{145}$. The $^{235}_{91}$Pa$_{144}$ ground state band[5] is probably $\frac{1}{2}^{-}$[530] as it is in the lighter odd-A Pa nuclei. The $I=\frac{1}{2}^{-}$ level in ^{231}Pa and ^{233}Pa is 9.2 keV and 6.6 keV, respectively, above the $I=\frac{3}{2}^{-}$ ground state. Thus the γ-ray spectra of these two nuclei show several doublets with corresponding ΔE values. However, no such doublets were observed in the ^{235}Th γ-ray spectra. The energies of the levels in the $K=\frac{1}{2}$ rotational band are given by the Bohr-Mottelson formula

$$E_{\rm rot}(K=\tfrac{1}{2},I)=\frac{\hbar^2}{2\mathscr{I}}I(I+1)+(-1)^{I+1/2}(I+\tfrac{1}{2})a_c \ ,$$

where a_c is the Coriolis decoupling parameter. Its value may strongly affect the relative positions of the band members. For $^{231}_{91}$Pa$_{140}$ and $^{233}_{91}$Pa$_{142}$, $a_c\leq -1$, and therefore the $I=\frac{3}{2}^{-}$ level is below the $I=\frac{1}{2}^{-}$ level. With in-

TABLE I. Gamma-ray energies E_γ and relative intensities I_γ following decay of 7.3-min ^{235}Th. Rough absolute intensities can be obtained by normalizing to 2.0 at 417.0 keV.

E_γ (keV)	I_γ	E_γ (keV)	I_γ
174.8(2)[a]	7.6(1.1)	704.0(2)	27.1(1.7)
292.9(1)	14.8(1.2)	708.3(2)	25.8(1.7)
406.1(1)[a]	14.9(1.4)	727.2(2)	43.3(2.0)
417.0(1)	100	729.5(2)	22.8(1.7)
450.4(2)	14.8(3.9)	737.0(5)	10.1(2.9)
468.7(2)	24.8(1.6)	747.8(2)	21.5(1.6)
484.2(2)[a]	19.4(4.0)	837.8(2)[a]	13.7(1.4)
644.9(2)[a]	28.2(2.8)	932.8(2)[a]	21.0(1.6)
696.1(2)	32.1(1.8)		

[a]Not placed in decay scheme.

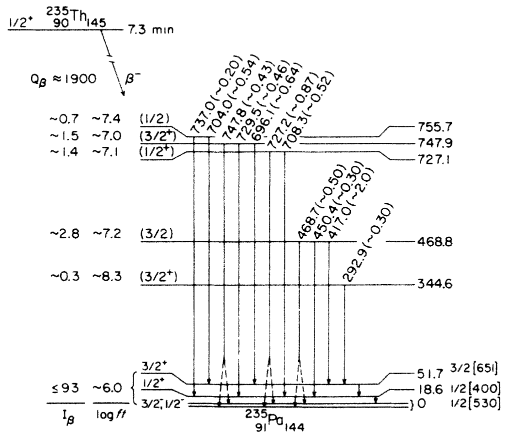

FIG. 2. Partial decay scheme proposed for decay of 7.3-min ^{235}Th. Estimated absolute intensities of the γ rays are shown in parentheses (see the text). The two lowest levels are not resolved; $\Delta E \leq 0.2$ keV.

creasing deformation Coriolis coupling weakens and therefore $|a_c|$ decreases. When $a_c \rightarrow 1$, $\Delta E[(I=\frac{3}{2})-(I=\frac{1}{2})]\rightarrow 0$. This seems to be the situation for $^{235}_{91}$Pa$_{144}$ where no γ-ray doublets were observed within the experimental limit of $\Delta E \leq 0.2$ keV. A value of $a_c \approx -1$ for ^{235}Pa is compatible with the observed $a_c = -1.46$ for ^{231}Pa and $a_c = -1.38$ for ^{233}Pa. The trend of decreasing a_c is an indication of increasing deformation of $_{91}$Pa nuclei as the neutron number increases from 140 to 144. The possibility cannot be excluded that in ^{235}Pa the $I=\frac{1}{2}$ level is the lower one, and thus it may be the ground state rather than the $I=\frac{3}{2}$ level.

All excited levels of ^{235}Pa populated directly by allowed or first-forbidden β transitions ($\log ft < 8$) must have $I=\frac{1}{2}$ or $\frac{3}{2}$. The level at 18.6 keV can be identified with the 19 ± 3 keV level observed in the (t,α) experiments[11] and assigned to the $\frac{1}{2}^+[400]$ rotational band with $I^\pi=\frac{1}{2}^+$. However, the level at 51.7 keV is probably not identical with the 55-keV level reported[11] in ^{236}U(t,α) and described as $\frac{7}{2}^-(\frac{1}{2}^-[530])$. The 51.7-keV level is populated from higher levels, above 300 keV, which do not feed the $\frac{3}{2}^-(\frac{1}{2}^-[530])$ ground state. Therefore the structure of this 51.7-keV level must be different from that of the ground state band. For similar reasons it is also different from the $\frac{1}{2}^+[400]$ rotational band based on the 18.6-keV level. The most probable assignment is to $\frac{3}{2}^+[651]$. Higher levels in ^{235}Pa (300–800 keV) most probably have $I=\frac{1}{2}$ or $I=\frac{3}{2}$ and are formed from collective states of an even-even core ($K=0^+$, $K=0^-$, $K=2^+$, . . . ,) combined with an odd proton in one of the above-mentioned Nilsson orbitals: $\frac{1}{2}^-[530]$, $\frac{1}{2}^+[400]$, and $\frac{3}{2}^+[651]$.

A very large number of levels in ^{235}U has been identified[5] in numerous reaction and decay studies; yet in the present work no significant β decay from ^{235}Pa was observed to any of these levels above 13 keV despite Q_β of ~ 1400 keV. In view of this result, it is useful to consider the effects of Nilsson asymptotic selection rules on the probability of β decay in deformed nuclei. An especially interesting situation occurs when β transitions populate two different Nilsson levels with identical asymptotic quantum numbers N, n_z, Λ, and with $\Delta\Omega=1$. In such cases the selection rules are identical for both β transitions and therefore the matrix elements will be similar. Such pairs of levels with relatively small ΔE were observed in many odd-A deformed nuclei:

$$Z=37,39:\ p\tfrac{1}{2}^-[301] \text{ and } p\tfrac{3}{2}^-[301]\ ,$$

$$Z=65-71:\ p\tfrac{1}{2}^+[411] \text{ and } p\tfrac{3}{2}^+[411]\ ,$$

$$N=93-97:\ n\tfrac{1}{2}^-[521] \text{ and } n\tfrac{3}{2}^-[521]\ ,$$

$$N=141-145:\ n\tfrac{1}{2}^+[631] \text{ and } n\tfrac{3}{2}^+[631]\ .$$

They are usually located on both sides of the Fermi surface, one of them a *particle* state and the other a *hole* state. In some of these nuclei both levels are populated by direct β decay, and the $\log ft$ values are known. However the analyses of such pairs of β transitions are usually complicated by pairing effects. According to Chasman *et al.*,[12] $1/(ft)_{\rm obs}=[1/(ft)_{\rm s.p.}]p_\beta^2$, where $(ft)_{\rm s.p.}$ is related to the matrix element for single-particle β transitions and $p_\beta^2 \approx 0.6-1.0$ for: β^- decay of odd-N nuclei and β^+, ϵ decay of odd-Z nuclei to *particle* states; and β^- decay of odd-Z nuclei and β^+, ϵ decay of odd-N nuclei to *hole* states. For the other types of transitions p_β^2 is usually small, $\ll 1$, and its value is very sensitive to details of the nuclear structure. Thus a clear cut analysis can be made only for the cases when p_β^2 is near 1.0 and when the $\log ft$ values are known. At present, these conditions are met only for β decay of ^{233}Pa$_{142}$ to ^{233}U$_{141}$ and ^{235}Pa$_{144}$ to ^{235}U$_{143}$. The β transitions (unhindered first forbidden) are the following: $\frac{1}{2}^-[530]\rightarrow\frac{1}{2}^+[631]$ and $\frac{1}{2}^-[530]\rightarrow\frac{3}{2}^+[631]$. In ^{235}U the particle level $\frac{1}{2}^+[631]$ is much closer to the Fermi surface ($E=0.08$ keV) than in ^{233}U ($E=399$ keV). Therefore p_β^2 is expected to be larger in ^{235}Pa than in ^{233}Pa, and $\log ft$ should be smaller. This agrees with the observed $\log ft$ values: 7.1 for ^{233}Pa and ~ 6.0 for ^{235}Pa. However for the $\frac{3}{2}^+[631]$ hole levels in ^{233}U and ^{235}U, $p_\beta^2 \approx 1.0$ and a normal $\log ft$ of 6–7 is expected for both β transitions. For ^{233}Pa this is true, with $\log ft = 7.2$, but for ^{235}Pa the analogous $\log ft > 9.5$. Thus this β transition is hindered by a factor of 100, contrary to the prediction of the Nilsson asymptotic selection rules. The reason for this is not clear.

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