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The α-decay of ^{236}Pu to ^{232}U

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Abstract

The γ-spectrum following the α-decay of ^{236}Pu was reinvestigated using a high resolution HPGe detector. The energy and intensity of 26 γ-transitions were measured accurately; 21 of these were observed for the first time. All γ-rays were placed in a ^{232}U level scheme accounting for 13 levels of which 8 are reported for the first time. The ground state rotational band was found to be fed up to spin $I^{\pi}=8^+$ and the $K^{\pi}=0^-$ octupole band to be fed up to $I=5$.

1. Introduction

^{232}U nuclear levels can be fed by ^{232}Np electronic capture or by ^{232}Pa β-decay [1]. Very few studies have been devoted to measurement of the ^{236}Pu α-decay, owing to the weakness of the sources available; the main study was that of Lederer [2] who measured both the γ-spectrum and α–γ coincidences.

2. Radiochemical procedure

^{236}Pu wa obtained by proton irradiation of ^{238}U targets (greater than 99.8% purity) at the isochronous Cyclotron (Orléans). The beam intensity was about $I=20$ μA and the time for each irradiation was between 60 and 90 h. Ten irradiations of ^{238}U targets were performed over a period of 15 months. At $E_p=34$ MeV, the ^{238}U(p,3n) reaction leads to ^{236}Np^{m} which generates ^{236}Pu; interferent reactions were ^{238}U(p,4n) ^{235}Np and ^{238}U(p,n) ^{238}Np $\beta\rightarrow$ ^{238}Pu and the fission reaction leading to the main part of the total activity. After a 2 month cooling time, the irradiated uranium targets were dissolved in hot concentrated HNO_3 and the solution adjusted to 7 M HNO_3 was loaded onto a Dowex 1 X-8 column where Pu^{4+}, Np^{4+} and Th^{4+} were fixed, while UO_2^{2+} passed through. Thorium was eluted by 10 M HCl and Np and Pu isotopes were recovered together with 2 M HCl. The Np–Pu separation was carried out, in 7 M HNO_3, on a small Dowex 1 X-8 column thermostated at 60 °C. Plutonium was eluted as Pu^{3+} with an 8 M HCl, 0.05 M HI solution. Unfortunately, the ingrowth of ^{232}U and ^{228}Th daughters and their daughters was significant in the first isolated plutonium fractions. Just before the final source preparation, all Pu fractions were evaporated to dryness, redissolved in 10 M HCl with two drops of concentrated HNO_3, and loaded onto a small anion exchange column. Only traces of tetravalent and pentavalent fission products (1 ppm) remained in the final source. A ^{236}Pu source of 1 cm^2 was prepared on a platinum disk (0.01 mm thickness) by electrodeposition from NH_4Cl medium [3]. The final activity, measured on aliquots by α-spectrometry, was (74 ± 8) MBq.

3. γ-Spectrometry and measurements

The γ-ray spectrometer consisted of a 40% relative efficiency coaxial HPGe detector with an energy resolution full width at half-maximum of 1.75 keV on the 1.33 MeV γ-line of ^{60}Co. This detector was coupled to a 8192 channel multichannel analyser (EG & G Ortec). The spectrometer was calibrated in energy and efficiency using multigamma sources such as ^{152}Eu, ^{133}Ba and ^{207}Bi.

γ-spectrum measurements were taken in less than 10 days after the source preparation. To enhance the high energy counting rate without degrading the resolution, we inserted between the source and the detector,

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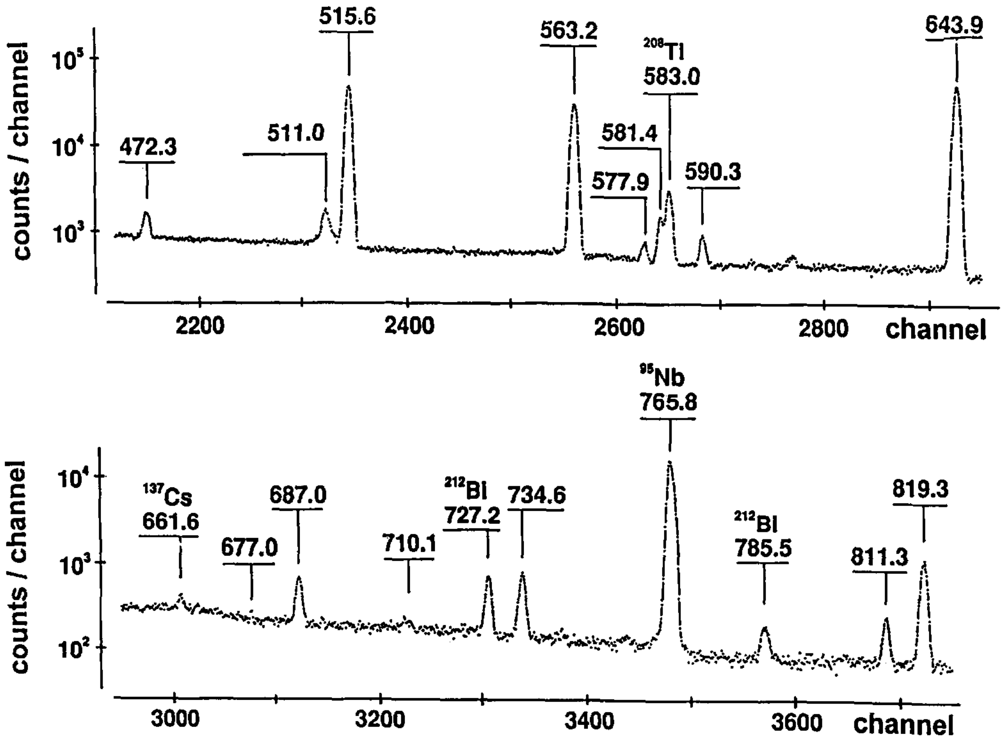

Fig. 1. Part of the γ-spectrum following the ^{236}Pu α-decay. The energies are in keV. γ-lines of ^{212}Bi and ^{208}Tl are due to the in-growth of the ^{232}U daughters in the electrodeposited ^{236}Pu source. Traces of fission product (less than 1 ppm) ^{95}Nb are also shown.

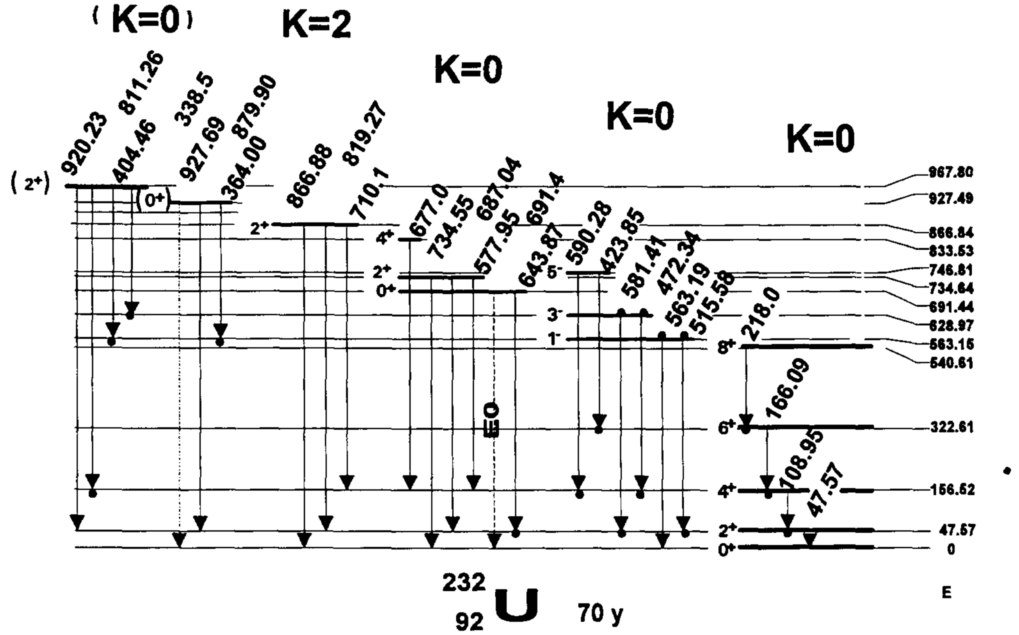

Fig. 2. ^{232}U level scheme fed in the α-decay of ^{236}Pu. Energies are given in keV. Observed γ–γ coincidences are indicated by heavy dots. The intensity of the E_0 transition of 691.4 keV was calculated from its branching ratio from ^{232}Pa β-decay [6].

a 1.5 mm thick lead sheet covered with a 1 mm copper sheet. Seven counting runs of about 15 h duration were carried out with this configuration. The ^{236}Pu γ-spectrum recorded is shown in Fig. 1, in which we can identify traces of fission products ^{95}Nb, ^{95}Zr and ^{125}Sb. Table 1 summarizes the data obtained in this analysis and compares them with the earlier work of Lederer [2]. Our intensity values were normalized allowing a ground state branching ratio equal to $(69.26 \pm 0.45)\%$ [1]. Twenty-six γ-transitions are reported in this study, 21 of which are new with respect to the study of Lederer [2].

TABLE 1. Energy and intensity of the γ-transitions following the α- decay of ^{236}Pu; uncertainties in parenthesis are given on the last digits

Present work		Lederer [2]	
E_γ (keV)	I_γ (%)	E_γ (keV)	I_γ (%)
47.57 (2)	0.065	47	0.031
108.95 (2)	0.0225	110	0.012
166.09 (5)	7.35 (2)×10^{-4}	165	6.6×10^{-4}
218.0 (1)	8.4 (1)×10^{-6}		
338.5 (1)	7.2 (1)×10^{-6}		
364.00 (10)	1.09 (15)×10^{-5}		
404.46 (10)	5.5 (1)×10^{-6}		
423.85 (20)	6.3 (1)×10^{-7}		
472.34 (10)	2.5 (2)×10^{-6}		
515.28 (2)	1.63 (5)×10^{-4}	520	1.7×10^{-4}
563.19 (2)	1.14 (4)×10^{-4}	570	1×10^{-4}
577.95 (10)	1.2 (2)×10^{-6}		
581.41 (10)	4.1 (2)×10^{-6}		
590.28 (10)	1.8 (1)×10^{-6}		
643.87 (3)	2.25 (9)×10^{-4}	640	2.4×10^{-4}
677.0 (2)	9.5 (4)×10^{-8}		
687.04 (10)	2.3 (1)×10^{-6}		
710.1 (3)	3.2 (1)×10^{-7}		
734.55 (10)	3.08 (13)×10^{-6}		
811.26 (20)	9.4 (1)×10^{-7}		
819.27 (10)	6.0 (2)×10^{-6}		
866.88 (10)	4.9 (3)×10^{-6}		
879.90 (10)	2.1 (1)×10^{-6}		
920.23 (20)	9.6 (1)×10^{-7}		
927.69 (20)	3.6 (4)×10^{-7}		

γ–γ coincidence measurements were taken using an experimental set-up consisting of three coaxial HPGe detectors of 20% efficiency and a planar HPGe detector of 20 cm^2 area. Each detector was positioned at 90° to its neighbor and the electrodeposited ^{236}Pu source was placed at the center of this set-up. Coincident events (γ–γ–t) were recorded in the event-by-event mode on magnetic tapes [4]; details of the hardware circuitry have been described elsewhere [5].

4. Discussion

The revised ^{232}U level scheme, shown in Fig. 2, is based on the present work and was built using sum relationship as well as γ–γ coincidence relationships analyzed out line from data obtained with the four-detector setup. All γ-transitions measured in this work were placed unambiguously in the level scheme. Their total intensities were calculated from values of γ intensities (Table 1) corrected for internal conversion, using the theoretical ICC values of Rösel *et al.* [7]. The total transition intensity is expressed in per cent of the α-decay.

First we discuss the ground state rotational band (g.s.). Levels with I^π up to 6^+ were known from the previous study of Lederer [2]. From the direct γ-spectrum and γ–γ coincidence relationships [4], we identified here the 218 keV γ-line as the $8^+ \to 6^+$ E_2 transition.

The $KI^\pi = 00^+$ band-head at 691.4 keV was suggested to be fed in the ^{236}Pu α-decay by Lederer [2] according to α–γ coincidence data. A strong γ-line of 643.9 keV was newly found here in the direct spectrum. The α-branching to this level was calculated taking into account the E_0 component of 691.4 keV of which intensity was deduced from its branching ratio from the ^{232}Pa β-decay [6]. The $I^\pi = 2^+$ member of this band was identified here from good energy relationships of the transitions of 734.55, 687.04 and 577.95 keV, interpreted as de-exciting it to the respective $I^\pi = 0$, 2 and 4^+ members of the g.s. band.

The α-feeding of the 866.8 keV ($KI^\pi = 22^+$) level was observed for the first time; it de-excites by γ-transitions of 866.88, 819.27 and 710.1 keV to the respective $I^\pi = 0$, 2 and 4^+ states of the g.s. band with branching ratio values 0.62:1.0:$<$0.1, in good agreement with those deduced from the ^{232}Pa β-decay [6].

We also have the $K^\pi = 0^-$ band at (563.15±0.20) keV. The feeding of the $I^\pi = 3^-$ member is observed for the first time in the ^{236}Pu decay and this level de-excites by the 581.41 and 472.34 keV transitions to the respective $KI^\pi = 02$ and 04^+ levels of the g.s. band. The $I^\pi = 5^-$ member of this band, already detected in the ^{230}Th (α, 2nγ) reaction [8], was observed here on the basis of the two γ-transitions of 590.3 and 423.8 keV, interpreted as de-exciting it to the respective $KI^\pi = 04$ and 06^+ states of the g.s. band.

A new (927.5±0.2) keV state is proposed to account for the γ-transition of 364.0 keV and the good sum relationships of the new γ-transition of 879.9 keV interpreted as de-exciting to the $KI^\pi = 02^+$ state of the g.s. band. According to this de-excitation pattern as well as the low hindrance factor value (HF = 14) [5], this level might be $I^\pi = 0^+$.

From our γ-intensity values we calculated the $B(E_1)/B(E_2)$ reduced probability ratio using the formula:

$$B(E_1;\, KI^+ \longrightarrow K'I'^-)/B(E_2;\, KI^+ \longrightarrow K''I''^+) = I_\gamma(E_1)\, E_\gamma(E_2)^5\, 0.03/[I_\gamma(E_2)\, E_\gamma(E_1)^3\, (197.1)^2] \quad (1)$$

where the energies E_γ are in mega-electronvolts; assuming an E_1 multipolarity for the γ-transition of 364 keV we obtained a $B(E_1)/B(E_2)$ ratio equal to $(4.4 \pm 0.6) \times 10^{-5}$ fm^{-2}, that can be compared with the Weisskoff estimate $B(E_1)_W/B(E_2)_W = 3.0 \times 10^{-2}$ fm^{-2}. If we assume that the E_2 de-excitations connecting this state to the g.s. members have $B(E_2) = 1$–2 W.u., we obtain a $B(E_1)$ of 1.5–3×10^{-3} W.u.

The new (967.8 ± 0.5) keV level allows the placement of four new γ-transitions of 920.23, 811.26, 404.46 and 338.58 keV, interpreted as de-exciting it to the respective $I^{\pi}=02^{+}$ and 00^{+} states of the g.s. band and to the $I^{\pi}=01^{-}$ and 03^{-} levels. Its spin will be restricted to $I^{\pi}=2^{+}$ or $I^{\pi}=3^{-}$. However, the low HF value, 6.7, deduced for this level [5] favors a 2^{+} assignment. It could be the 2^{+} member of a second excited $K^{\pi}=0^{+}$ band based on the 927.7 keV state.

The moment of inertia, $I=74.5$ MeV $\hbar^{-2}$, calculated with this assumption, is only 18% higher than that of the g.s. band. In the ^{234}U neighboring isotope a second excited $K^{\pi}=0^{+}$ band lies at 1044.5 keV with the 2^{+} member at 1085.3 keV and respective hindrance factors HF = 6.8 and 4.7.

The respective values $B(E_1;\ K_2{}^{+}\rightarrow 03^{-})/B(E_2;\ K_2{}^{+}\rightarrow 04^{+})=(5.4\pm0.9)\times10^{-5}$ fm^{-2} and $B(E_1;\ K_2{}^{+}\rightarrow 01^{-})/B(E_2;\ K_2{}^{+}\rightarrow 04^{+})=(4.4\pm0.9)\times10^{-5}$ fm^{-2} are consistent and then imply an average $B(E_1)$ of $(1.5\text{–}3)\times10^{-3}$ W.u. This enhancement of the E_1 strength can be compared with those obtained in lighter thorium nuclei and faster than the rates in nuclei heavier than ^{228}Th [9]. Such a characteristic suggests that this $K^{\pi}=0^{+}$ band could contain strong two-octupole phonon components.

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