Levels of ^{232}U fed in ^{236}Pu α decay

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The γ spectrum following the α decay of ^{236}Pu has been reinvestigated with a high-resolution HPGe detector. Accurate energies and intensities are reported for 26 γ transitions, of which 20 were observed for the first time. A decay scheme is constructed using the Ritz combination principle, $\gamma\gamma$ coincidence data, and previously known data from nuclear reactions and from ^{232}Pa β^- decay. We observe feeding of the ground-state rotational band up to spin 8, the β vibrational band to spin 4, the $K = 0$ octupole vibrational band to spin 5, and the bandhead of the γ vibrational band. New states of ^{232}U at 927.3 and 967.7 keV, populated with low α-decay hindrance factors, are assigned as members of a second-excited $K^\pi = 0^+$ band. The ratio of $E1/E2$ transitions in the decay of these states suggests that the $E1$ transitions to members of the $K = 0$ octupole band may be rather fast ($\approx 10^{-3}$ Weisskopf units). Systematics of hindrance factors for α decay to vibrational states are presented.

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I. INTRODUCTION

^{232}U is a nuclide at the borderline of the mass region ($220 \leq A \leq 230$), in which stable reflection-asymmetric octupole deformations occur [1]. Its neighbor ^{234}U is a typical quadrupole-deformed nucleus [2], whereas ^{228}Th has a much lower lying $K^\pi = 0^-$ band, suggestive of reflection asymmetry [3]. Excited states of ^{232}U have been studied in ^{232}Pa β^- decay [4–6], ^{232}Np electron capture decay [7], and the reactions ^{230}Th$(\alpha, 2n\gamma)$ [8,9] and ^{232}Th$(\alpha, 4n\gamma)$ [10–12]. There have been few measurements of ^{236}Pu α decay, due to the weakness of the sources available. The main studies are those of Hummel, who investigated the α-particle spectrum and coincident low-energy γ rays [13], and Lederer, who measured $\alpha\gamma$ and αe^- coincidences [14].

II. EXPERIMENTAL DETAILS

A. Radiochemical separation of ^{236}Pu

^{236}Pu was produced by 34-MeV proton irradiation of 3-mm-thick, 38-g depleted uranium targets ($> 99.8\%$ ^{238}U) at the Isochronous Cyclotron (CERI Orléans). With a view to obtaining a very strong source needed for our investigation on exotic (^{28}Mg) emission of ^{236}Pu, we performed ten irradiations over a period of 15 months, of duration between 60 and 90 h, with a beam current of approximately 20 μA.

The proton energy and target thickness were chosen to optimize the production of ^{236}Np^m by the reaction ^{238}U$(p, 3n)$; 22.5-h ^{236}Np^m decays to ^{236}Pu by a 50% β^- branch. Other activities produced under conditions of the irradiation include 396-d ^{235}Np, 2.12-d ^{238}Np, its 87.7-yr ^{238}Pu daughter, and fission products, which constitute most of the total activity prior to chemical purification.

After a cooling time of about 2 months, the uranium targets were dissolved in hot, concentrated HNO_3. The resulting solution, adjusted to $7M$ HNO_3 and a $UO_2{}^{2+}$ concentration of about 150 g/l, was loaded onto a column (height 20 cm, diameter 1 cm) containing Dowex 1×8 anion-exchange resin, on which Pu^{4+}, Np^{4+}, and Th^{4+} adsorb while $UO_2{}^{2+}$ passes through. Thorium was eluted with a $10M$ HCl solution, and neptunium and plutonium were eluted together with a $2M$ HCl solution. Plutonium was separated from neptunium on a small anion-exchange column (height 6 cm, diameter 0.3 cm) thermostatted at 60 °C. After adsorption from a $7M$ HNO_3 solution, which eliminated last traces of uranium, a reducing solution consisting of $0.05M$ HI in $8M$ HCl eluted the plutonium as Pu^{3+}. ^{236}Pu isolated from the ten targets was combined to make the final source.

Although the ^{232}U daughter and ^{228}Th granddaughter of ^{236}Pu have relatively long half-lives (71.7 and 1.9 yr, respectively), the short-lived descendants ^{212}Bi, ^{212}Pb, and ^{208}Tl, which emit intense γ rays, appear quickly in the spectrum with intensities comparable to the intensities of the weak ^{236}Pu γ rays. In order to minimize

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this interference, the daughters were removed just before the final source preparation. The plutonium fraction was evaporated to near dryness, redissolved in 10M HCl with 2 drops of concentrated HNO_3, and loaded onto a small anion-exchange column. ^{228}Th was eluted with 10M HCl, followed by 8M HNO_3 to remove ^{232}U and iron impurities. Finally, plutonium was recovered in less than 1 cm^3 of 2M HCl. Only traces of the tetravalent and pentavalent fission products (^{95}Zr, ^{95}Nb, and ^{125}Sb) remained in the final source, their activity amounting to less than 10^{-4}% of the plutonium activity.

A ^{236}Pu source of 1 cm^2 area was prepared on a platinum disk (0.01 mm thickness) by electrodeposition from an NH_4Cl medium. A more detailed account of the separation scheme will be published elsewhere [15]. The final activity, measured by α-spectrometric assay of aliquots with a Si(Au) detector and a 2π ionization chamber, was 74 ± 8 MBq.

B. Spectrometers

The spectrometer for γ-ray singles measurements was a 40% relative efficiency coaxial n-type HPGe detector (Canberra) with an energy resolution (FWHM) of 1.75 keV for the 1333-keV γ ray of ^{60}Co. Spectra were recorded with an 8192-channel multichannel analyzer (EG&G Ortec). To reduce background contributions the

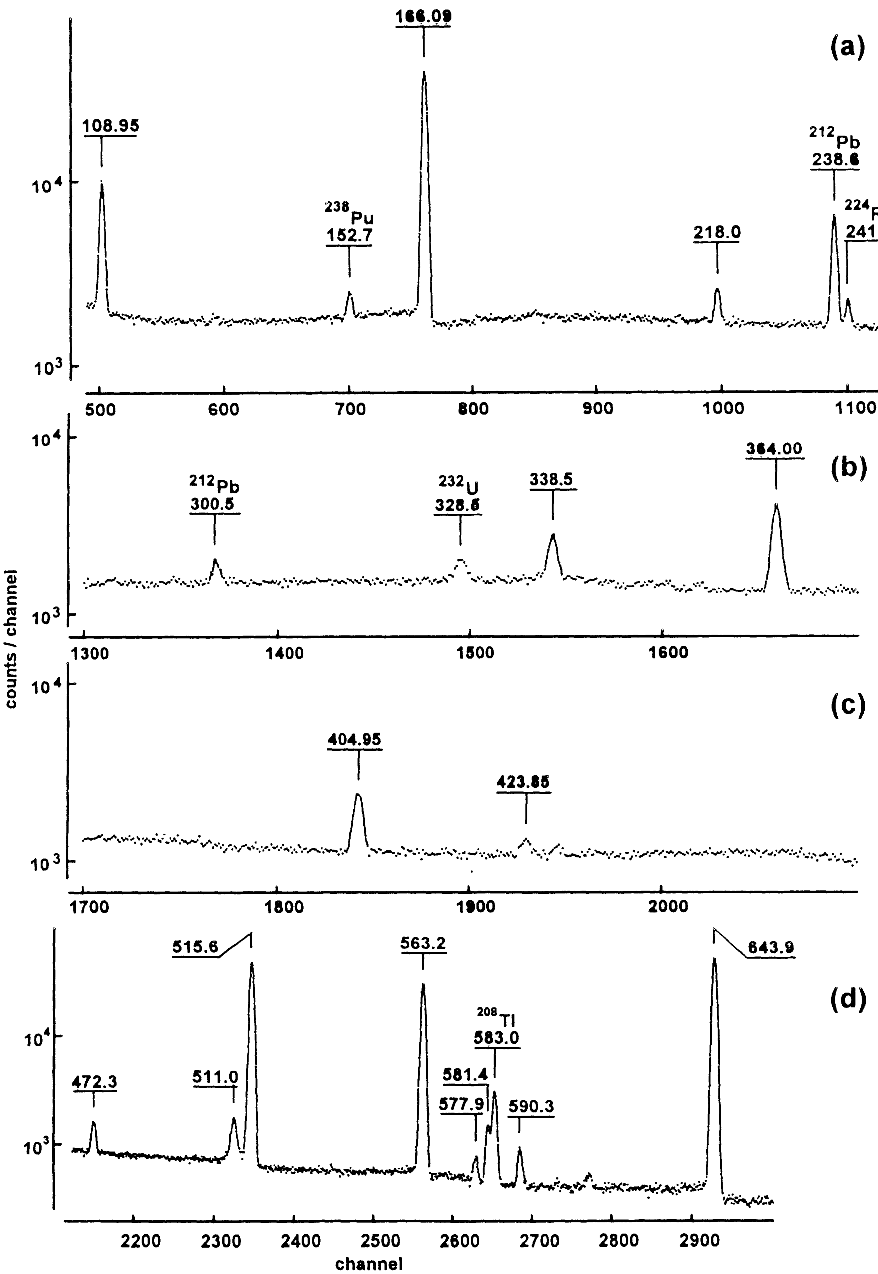

FIG. 1. The γ spectrum following ^{236}Pu α decay. Energies are in keV. γ rays of ^{212}Bi and ^{208}Tl are mainly due to the ingrowth of the ^{232}U daughters in the source. Traces of the fission product ^{95}Nb (< 1 ppm) are also evident.

detector head was inserted in a lead shield of 5 cm thickness internally covered with 2-mm-thick copper foils. The spectrometer was calibrated in energy and efficiency with multigamma standard sources such as ^{152}Eu, ^{133}Ba, and ^{207}Bi.

$\gamma\gamma$ coincidences were measured with three n-type coaxial HPGe detectors of 20% relative efficiency, placed at angles of 90°, 180°, and 270° with respect to a planar HPGe LEPS detector of 20 cm^2 area. The energy resolution was 1.8-1.9 keV (FWHM) at 1333 keV for the coaxial detectors, 0.50 keV at 122 keV for the LEPS detector. The electrodeposited source was located 5 cm from each detector. Events coincident between any pair of the four detectors were selected by a standard hardware circuitry (MIPRE system) and the information from TAC and ADC converters was stored event-by-event on magnetic tape.

III. MEASUREMENTS AND RESULTS

γ spectra were measured within 10 days after source preparation to reduce the contribution from ^{236}Pu descendants. To reduce the contribution of 47.57 and 108.95 keV γ rays, we inserted an absorber consisting of a 1.5-mm-thick lead sheet covered with a 1-mm copper sheet between the source and the detector when measuring the spectrum above 300 keV. Several counting runs of about 15 h duration were carried with this configuration and a source-to-detector distance of 10 cm. In this geometry the counting rate is low and summing effects are negligible. The spectrum recorded under these conditions is shown in Fig. 1.

The spectrum includes a broad peak (FWHM=7.6 keV) around 870 keV. When the electrodeposited source

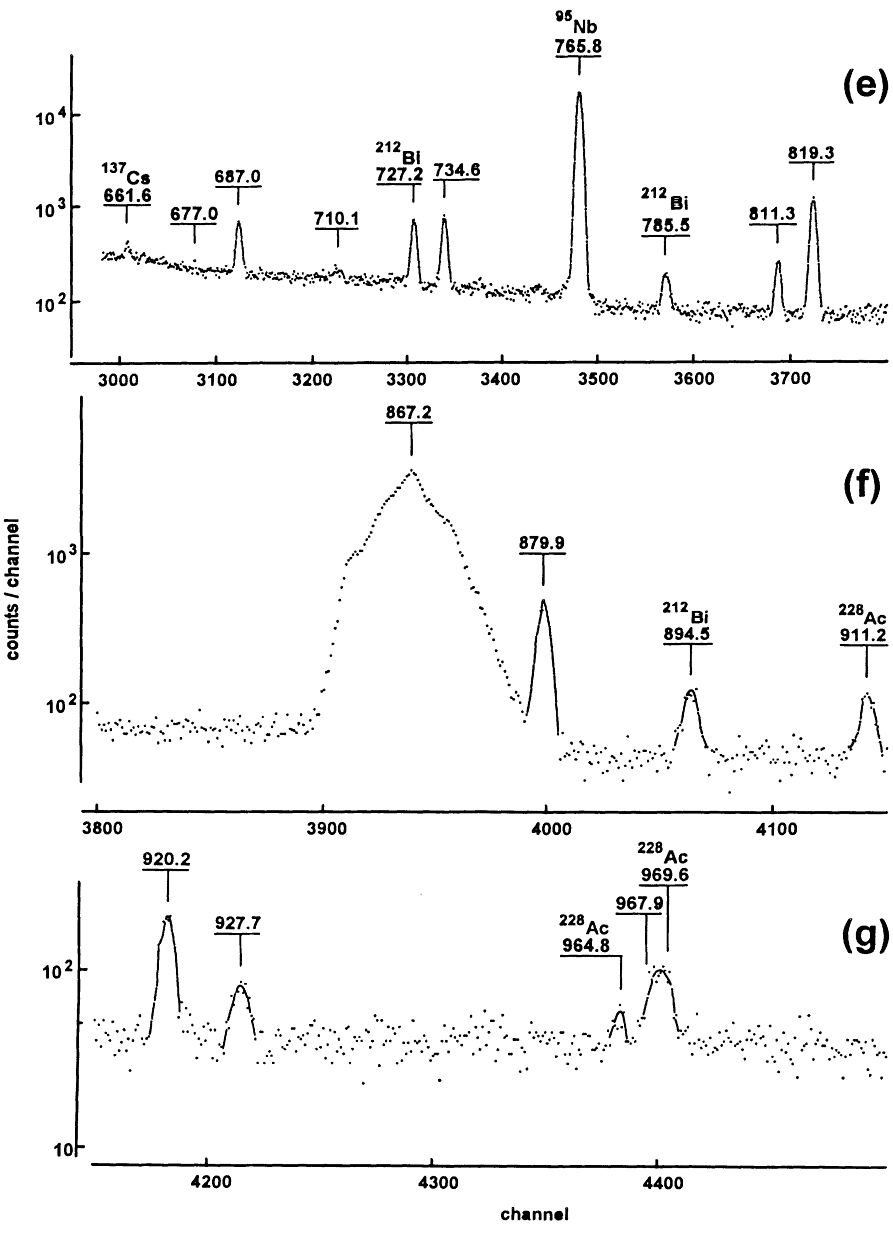

FIG. 1 *(Continued)*.

is replaced with a ^{236}Pu solution, the broad peak disappears, revealing three well-resolved photopeaks. The 870-keV peak is caused by the $^{14}N(\alpha,p)$ reaction on the nitrogen of the air surrounding the source, which populates the first excited state of ^{17}O at 870.8 keV [16]. The Doppler-broadened γ ray that de-excites this state masks the γ rays from the source.

Table I summarizes the data obtained from our analysis of the spectrum and compares them to previous work. γ rays from activities other than ^{236}Pu—descendants of ^{236}Pu and traces of the fission products ^{95}Zr, ^{95}Nb, and ^{125}Sb—are omitted from the table. Their well-known γ spectra are easily distinguished from the γ rays of ^{236}Pu. Our intensity normalization is based on a ground-state (g.s.) α branching of (69.14±0.33)% [17]. We assign 26 γ-ray transitions to the decay of ^{236}Pu, including 20 not previously seen in this decay scheme.

Coincidences were measured during a four-day counting period. A total of 6.3×10^5 $\gamma\gamma t$ events were collected. Spectra were analyzed off-line by setting gates on several γ-ray lines. Typical coincidence spectra are shown in Fig. 2, and Table II summarizes the results obtained with different gates.

IV. DISCUSSION

The revised ^{236}Pu decay scheme shown in Fig. 3 was constructed using data from the present work and previous data on ^{236}Pu decay, ^{232}Pa decay, ^{232}Np decay, and $(\alpha,xn\gamma)$ reactions. The observed γ rays, with the exception of the 927.69-eV transition as discussed below, are

TABLE I. Energies and intensities of γ rays following ^{236}Pu α decay.

Present work		Previous work	
E_γ (keV)	I_γ (%)	E_γ (keV)	I_γ (%)
47.57 (2)	0.065	47.65 (5)[a]	0.031[b]
108.95 (2)	0.0225	108.96 (5)[a]	0.012[b]
166.09 (5)	7.35 (2) $\times10^{-4}$	166.3 (2)[c]	6.6 $\times10^{-4}$ [b]
218.0 (1)	8.4 (1) $\times10^{-6}$	218.4 (1)[d]	
338.5 (1)	7.2 (1) $\times10^{-6}$		
364.00 (10)	1.09 (15) $\times10^{-5}$		
404.46 (10)	5.5 (1) $\times10^{-6}$		
423.85 (20)	6.3 (1) $\times10^{-7}$	424.3 (5)[e]	
472.34 (10)	2.5 (2) $\times10^{-6}$	472.390 (6)[f]	
515.58 (2)	1.63 (5) $\times10^{-4}$	515.607 (9)[f]	1.7 $\times10^{-4}$ [g]
563.19 (2)	1.14 (4) $\times10^{-4}$	563.197 (7)[f]	1.0 $\times10^{-4}$ [g]
577.95 (10)	1.2 (2) $\times10^{-6}$		
581.41 (10)	4.1 (2) $\times10^{-6}$	581.398 (8)[f]	
590.28 (10)	1.8 (1) $\times10^{-6}$	590.4 (5)[e]	
643.87 (3)	2.25 (9) $\times10^{-4}$	≈645[h]	2.4 (3) $\times10^{-4}$ [g]
677.0 (2)	9.5 (4) $\times10^{-8}$	676.5 (2)[i]	
687.04 (10)	2.3 (1) $\times10^{-6}$	687.0 (1)[j]	
		691.3 (1)[j] (E0)	3.5 (10) $\times10^{-4}$ [g]
710.1 (3)	3.2 (1) $\times10^{-7}$	710.1 (1)[j]	
734.55 (10)	3.08 (13) $\times10^{-6}$		
811.26 (20)	9.4 (1) $\times10^{-7}$		
819.27 (10)	6.0 (2) $\times10^{-6}$	819.187 (13)[f]	
866.88 (10)	4.9 (3) $\times10^{-6}$	866.760 (19)[f]	
879.90 (10)	2.1 (1) $\times10^{-6}$		
920.23 (20)	9.6 (1) $\times10^{-7}$		
927.69 (20)[k]	3.6 (4) $\times10^{-7}$		
967.9 (3)[l]	3.5 (8) $\times10^{-7}$		

[a] ^{232}Pa β^- decay [6].
[b] Reference [13].
[c] ^{232}Th$(\alpha,4n\gamma)$ [12].
[d] ^{232}Th$(\alpha,4n\gamma)$ [11].
[e] ^{230}Th$(\alpha,2n\gamma)$ [8,24].
[f] ^{232}Pa β^- decay [25].
[g] Reference [14].
[h] ^{232}Pa β^- decay [4].
[i] ^{230}Th$(\alpha,2n\gamma)$ [9].
[j] ^{232}Pa β^- decay [5].
[k] Not placed in the level scheme (see text).
[l] Distinct from the well-known 968.97-keV γ ray from ^{228}Ac decay [3], present in the background of the measurement room.

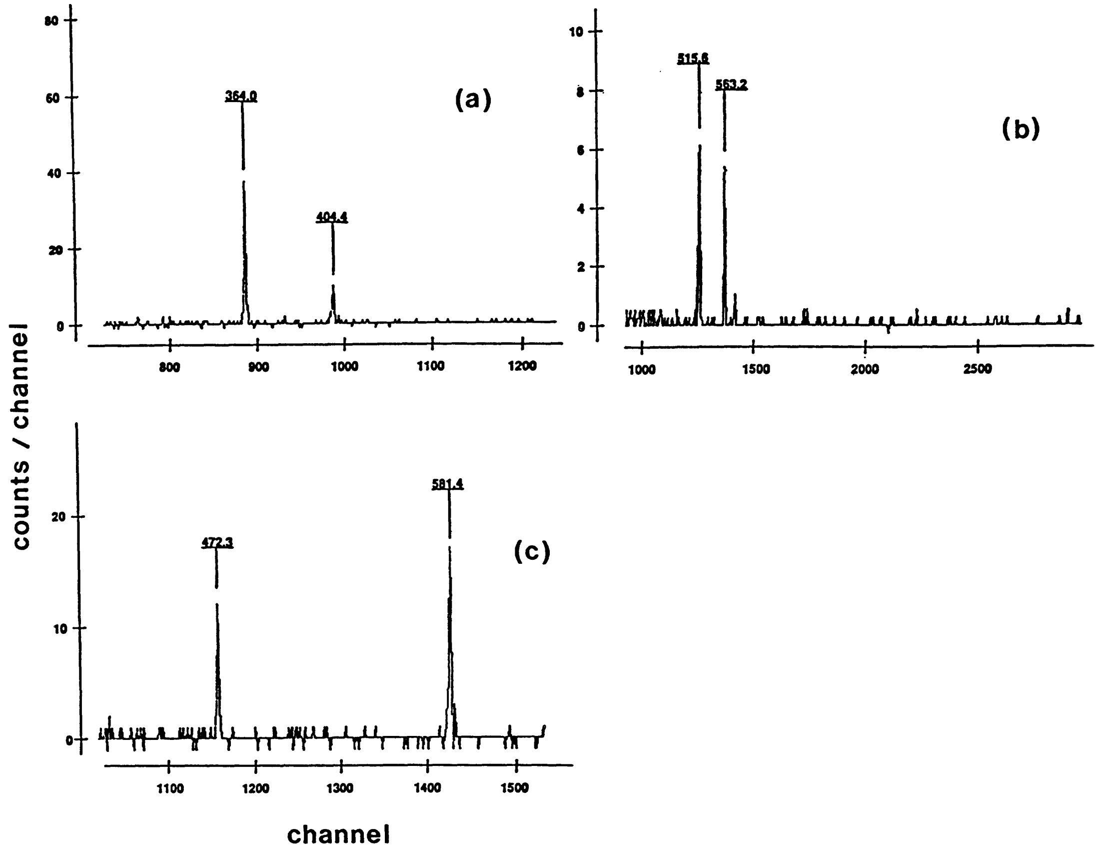

FIG. 2. Portions of the γ spectrum coincident with gates set on the transitions at (a) 563 keV, (b) 404 keV, (c) 338 keV. Energies are in keV.

placed in the level scheme on the basis of their energies and the coincidence results.

The α-transition intensities in Fig. 3 were calculated from the γ-ray intensities, converted to transition intensities with the use of theoretical internal conversion coefficients (ICC) of Rösel *et al.* [18]. Multipolarities adopted for this purpose are based on measured ICCs and the adopted spins of the initial and final states. Additional multipolarity assignments are derived from the γ-ray branching ratios. The derivation of these multipolarities and the calculation of intensities for unobserved γ rays and $E0$ transitions are described below. α-decay hindrance factors (HF) were calculated from the one-body spin-independent model of Preston [19].

The ground-state rotational band. Levels of the g.s. band through spin 20 are known from the ^{232}Th$(\alpha, 4n\gamma)$ reaction [10]. Levels through spin 6 have previously been observed in ^{236}Pu α decay [13]. Observation of the $8+ \to 6+$ transition establishes the α decay to the spin-8 member of the band at 540.70±0.12 keV with an intensity of $(1.3 \pm 0.2) \times 10^{-5}$%.

The $K = 0$ octupole vibrational band. This band is well established from the decay of ^{232}Pa [4]. States through spin 13 have been observed in the ^{232}Th$(\alpha, 4n\gamma)$ reaction [8]. Our measurements yield an improved energy for the spin-5 member: 746.79±0.13 keV. Lederer showed that α decay populates the bandhead with an intensity of $(2.7 \pm 0.3) \times 10^{-4}$% and set a limit of 1.5×10^{-5}% on the population of the spin-3 member [14]. We observe γ rays deexciting spin-1, -3, and -5 members of the band. The α feeding of the spin-1 state is $(2.6 \pm 0.1) \times 10^{-4}$%, in agreement with the previous value. The 3^- state appears to be fed only by a transition from the 967.66-keV state; direct α feeding, as estimated from the intensity balance, is $< 1 \times 10^{-6}$%. α decay feeds the 5^- state with an intensity $(2.5 \pm 0.1) \times 10^{-5}$. Thus, the $l = 5$ α transition is less hindered than the $l = 3$. Inversion of the α

TABLE II. Summary of the $\gamma\gamma$ coincidences following ^{236}Pu α decay.

Gate energy (keV)	Coincident γ-ray energy (keV)
47	108,166,515,643,811
109	47,166,472,590
166	109,218,423
338	472,581
364	515,563
404	515,563
472	109,338
515	364
563	364,404

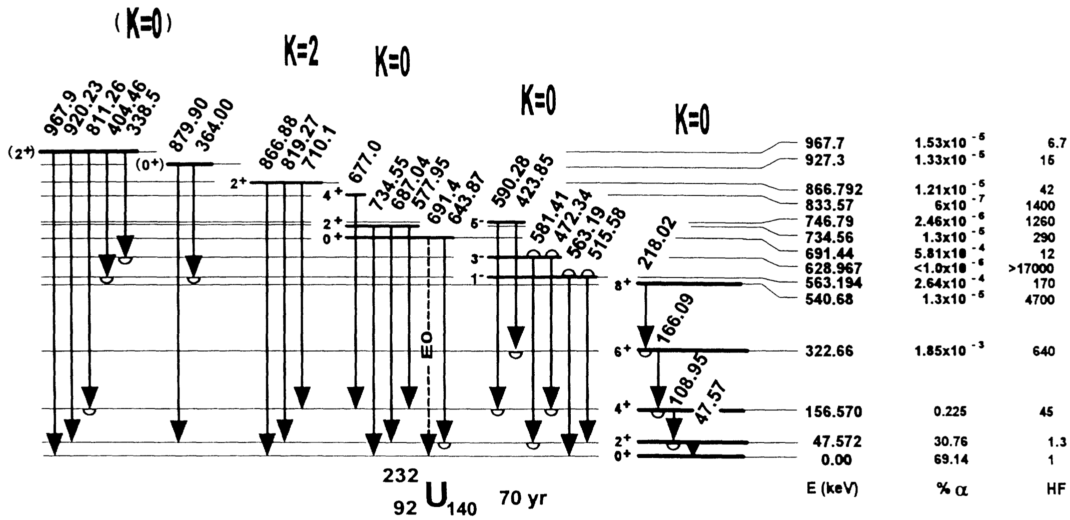

FIG. 3. ^{232}U levels fed in the α decay of ^{236}Pu. Energies are in keV. Observed $\gamma\gamma$ coincidences are noted by open semicircles. α branchings and hindrance factors are shown on the right. Level energies and J^π assignments are adopted values, based on the present results and previous experiments.

population of the 3^- and 5^- states also occurs in ^{232}U decay to ^{228}Th [20].

The β vibrational band (states at 691.44±0.05, 734.56±0.06, and 833.53±0.20 keV). Lederer observed α decay to the $0^{+'}$ β vibrational state with an intensity 5.9×10^{-4}% and set a limit of 2×10^{-5}% on a possible α branch to the spin-2 member of the band [14]. Our data confirm these results and provide evidence of α decay to the spin-2 and spin-4 members of the band. The α branching to the 691.44-keV 0^+ state, $(5.8 \pm 1.0) \times 10^{-4}$%, is calculated from our γ-ray intensities and the $E0$ intensity deduced from the α-e^- coincidence measurement [14]. γ-ray branching from the spin-2 member of the β vibrational band yields a mixing parameter $z_{\beta g} = -0.016 \pm 0.004$ and a pure $E2$ multipolarity for the 687.04-keV γ ray ($\delta^2 > 10$). α feeding of this state, $(1.3 \pm 0.1) \times 10^{-5}$%, is calculated from the measured γ-ray intensities and an $E0$(687.04 keV, $02^{+'} \to 02^+$) intensity of $(6.8 \pm 2.0) \times 10^{-6}$%, which we estimate by assuming that the ratio of $E0/E2$ strength is the same as for the $00^{+'}$ state.

α decay to the $04^{+'}$ state is inferred from the 677.0-keV ($04^{+'} \to 04^+$) γ ray. Transitions from the $04^{+'}$ state to the 02^+ and 06^+ states are masked by the 785.5-keV ^{212}Bi γ ray and annihilation radiation, respectively. From I_γ(677.0 keV) and the value of $z_{\beta g}$ calculated for the $02^{+'}$ state, the expected γ-ray intensities are $(3.2 \pm 0.3) \times 10^{-7}$% ($04^{+'} \to 02^+$) and $(1.7 \pm 0.2) \times 10^{-8}$% ($04^{+'} \to 06^+$). The calculated ($04^{+'} \to 02^+$) intensity is higher than the experimental limit of $\leq 1.6 \times 10^{-7}$% on the intensity of a possible ^{236}Pu γ ray at 786 keV. For estimation of the α feeding of the 833.5-keV $04^{+'}$ state, we adopt an intensity of 1.6×10^{-7}% for this transition, $\approx 2.7 \times 10^{-7}$% for the total intensity of the γ rays (photons) deexciting the $04^{+'}$ state. The calculated $E0$ intensity of the $04^{+'} \to 04^+$ transition is $(3.2 \pm 0.9) \times 10^{-7}$%, based on the assumption that the $E0/E2$ strength ratio the same as for the $00^{+'}$ state. Thus the calculated α feeding of the $04^{+'}$ state is 6×10^{-7}%.

The γ vibrational band. This band and its mixing with the β vibrational band have been characterized in detail in the decay of ^{232}Pa [4]. The 2^+ bandhead is populated with an α intensity of $(1.21 \pm 0.06) \times 10^{-5}$%. (The calculated feeding is corrected for the presence of weak transitions observed to deexcite the state in ^{232}Pa decay [4].) There is no evidence for population of the spin-4 member of the band at 970 keV; absence of an 814-keV γ ray ($24^+ \to 04^+$, $I_\gamma < 4 \times 10^{-8}$%) sets a limit of 5×10^{-8}% on the α feeding of this state.

States at 927.3±0.2 and 967.7±0.2 keV. These states are observed here for the first time. Their existence is firmly established by the γ-ray energies and the coincidence results. Although we lack multipolarities on which to base firm J^π assignments, there is strong evidence that these states are the spin-0 and -2 members of a $K^\pi = 0^+$ band: (1) Their deexcitation pattern suggests that they are members of the same band, and the 40-keV separation between the states favors the assignments 0^+-2^+. (If the states belong to the same band and are both populated directly by α decay, possible combinations of J^π values are limited to 0^+-2^+, 1^--3^-, 2^+-4^+, etc.) (2) The deexcitation pattern limits the choice of spin values. If the γ-ray transitions from the 967.66-keV state to spin-0, -2, and -4 members of the g.s. band are correctly placed, a 2^+ assignment for this state is nearly certain. (3) The α-decay hindrance factors are very low. For nuclei in this region, only the favored α transition and transitions to excited $K^\pi = 0^+$ states (and their analogs in odd-mass nuclei) have hindrance factors under 10. Low hindrance for the α transition to the 0^+ state and an even less hindered α transition to the spin-2 member of the band appear to be characteristic of second-excited $K^{\pi} = 0^{+}$ bands, based on two other cases in which analogous α transitions have been observed (the decay of ^{238}Pu and ^{242}Cm). However, the occurrence of a 927.7-keV γ ray is tentative evidence against the proposed assignments. If this is a ground-state transition from the 927.3-keV state, a 0^{+} assignment for the state must be ruled out. Because other evidence strongly favors the proposed assignments, we leave this transition unplaced; its assignment to the decay of ^{236}Pu needs to be verified and its placement determined.

Decay of the 927.3- and 967.7-keV states to members of the $K = 0$ octupole band suggests an interpretation of the band as a two-phonon octupole vibration. The ratio of the $E1$ intensities to the intensities of $E2$ transitions to the g.s. band, each expressed in Weisskopf single-particle units (W.u.), is 1.5×10^{-3}. If the $E2$ transitions have strengths of about one single-particle unit, then the $E1$ strengths are $\approx 10^{-3}$ W.u. This is comparable to the rates of the one-phonon→zero-phonon octupole-band transitions in lighter thorium nuclei and faster than the rates in nuclei heavier than ^{228}Th [8,21]. Interpretation of the $K^{\pi} = 0^{+''}$ band as a two-phonon octupole excitation is in agreement with the calculations of Ivanova *et al.*, which predict that this configuration will be the main component (80% squared amplitude) of the second-excited $K^{\pi} = 0^{+}$ band in ^{232}U, whereas the first-excited $K^{\pi} = 0^{+}$ band is predicted to be mainly a one-phonon β vibration (84%), with only 3% of the two-phonon octupole configuration [22].

Two-band mixing analysis of the γ-ray branching ratios from the 967.7-keV state to members of the g.s. band yields a mixing parameter $z_{\beta' g} = +0.025 \pm 0.007$ and an $E2/M1$ mixing ratio $\delta^2 = 1.3 \pm 0.4$ for the 920.23-keV $02^{+''} \rightarrow 02^{+}$ transition.

V. CONCLUSIONS

Experimental systematics of the hindrance factors for vibrational states are shown in Figs. 4(a)–4(c). For the $K = 0$ octupole vibrational band [Fig. 4(a)] the hindrance of the $l = 1$ alpha transition has a broad maximum, and HF($l = 3$) has a sharper maximum, around neutron number 140. Around $N = 132$ the $l = 1$ and $l = 3$ hindrance factors become very low, and there is a broad, deep minimum in the energy of the band around $N = 136$. This minimum, the low hindrance factors, and even lower hindrance factors for analogous transitions in odd-mass and odd-odd nuclei have been interpreted as evidence of a tendency toward stable reflection asymmetry [1]. It would be of interest to know whether the trend toward *higher* hindrance factors for $l = 5$ transitions continues below $N = 136$ and, if so, how the interpretation of these states might be affected.

Figure 4(b) shows the hindrance factors for β vibrational ($K^{\pi} = 0^{+'}$) bands. Hindrance factors for $l = 0$ transitions are low and nearly constant, whereas the $l = 2$ hindrance factors rise dramatically around $N = 140$. The

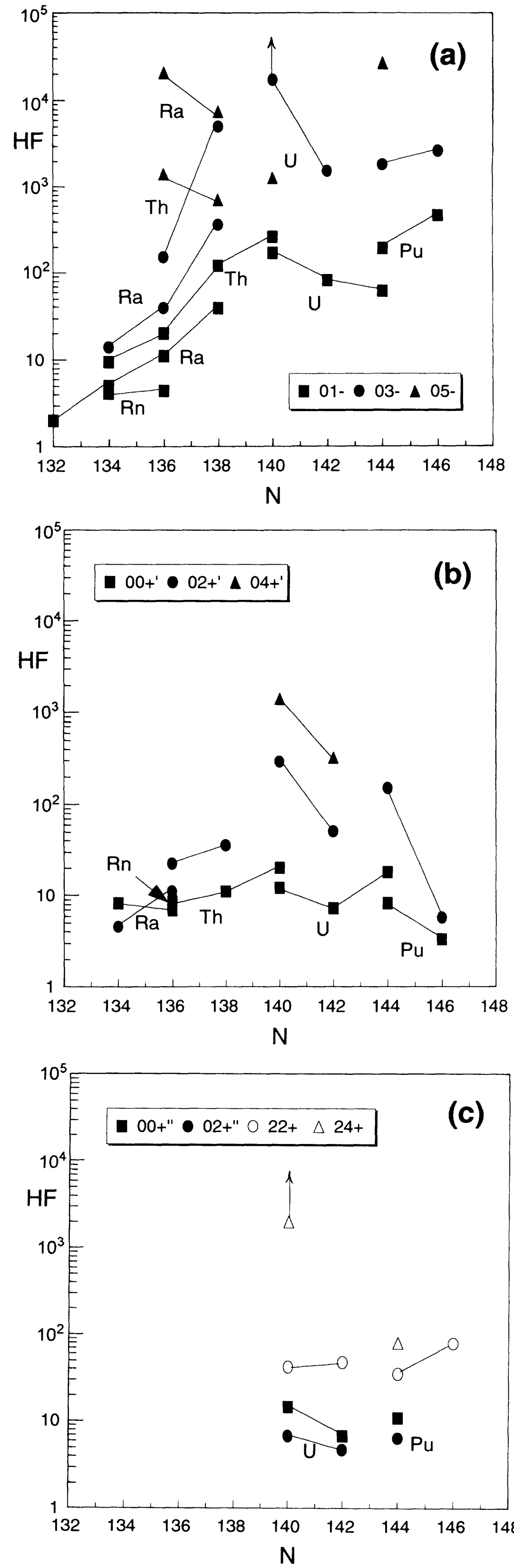

FIG. 4. Systematics of hindrance factors for α decay to vibrational states. The neutron number N and the element symbols refer to the daughter nucleus. (a) $K = 0$ octupole vibrational states, (b) β vibrational ($K^{\pi} = 0^{+'}$) states, (c) γ vibrational states and $K^{\pi} = 0^{+''}$ states.

$l = 4$ transitions are quite hindered and also appear to have a maximum near $N = 140$.

Figure 4(c) summarizes the known hindrance factors for α decay to γ vibrational bands and second-excited $K^\pi = 0^+$ bands. HFs for the γ vibrational bandhead (four cases) are nearly constant and rather large compared to $l = 2$ transitions to the g.s. band. As noted above, HFs for the second-excited $K^\pi = 0^+$ bands are low, and in all three cases the $l = 2$ transition is less hindered than the $l = 0$ transition.

Over 20 years ago Sandulescu and co-workers attempted to estimate the strengths of α-decay transitions to vibrational states with the use of Nilsson wave functions, pairing and quadrupole-quadrupole residual interactions, and the random phase approximation [23]. The growing body of data warrants a renewed effort to understand the systematics. In particular, the constancy of the hindrance factors for the $00^{+'}$, $00^{+''}$, $02^{+''}$, and 22^+ states, the surprisingly low HFs for the $02^{+''}$ states, and the large increase in the HFs for the spin-2 member and perhaps the spin-4 member of the β vibrational ($K^\pi = 0^{+'}$) band around $N = 140$ have not been satisfactorily explained.

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