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Identification and decay of ^{242}U and ^{242}Np

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The nuclides ^{242}U and ^{242}Np were discovered by chemical separations from targets of ^{244}Pu irradiated with 30–160 MeV neutrons followed by γ-ray and β-ray spectroscopy. The parent ^{242}U decays with $T_{1/2} = 16.8 \pm 0.5$ min; 14 γ rays ranging from 55.6 keV to 585.0 keV were assigned to its decay. The ^{242}Np daughter half-life is 2.2 ± 0.2 min and 39 γ rays following its decay have energies between 620.6 and 2370.5 keV. Q_{β^-} of ^{242}Np is 2.7 ± 0.2 MeV. A level scheme is proposed for ^{242}Pu in which the lowest 7 levels were previously described and 12 new levels are shown. The ground state of ^{242}Np is assigned $I^\pi = 1^+$ and 6 excited levels are proposed which accommodate most of the observed γ rays from decay of ^{242}U.

RADIOACTIVITY ^{242}U, ^{242}Np from ^{244}Pu$(n, 2pn)^{242}$U$\xrightarrow{\beta^-}$^{242}Np; radiochemistry, Ge(Li) and plastic detectors. ^{242}U: measured $T_{1/2}$, E_γ, I_γ; deduced E_{β^-}, I_{β^-}, $\log ft$, decay scheme, J, π, Nilsson state assignments. ^{242}Np: measured $T_{1/2}$, E_γ, I_γ, E_{β^-}, I_{β^-}; deduced $\log ft$, decay scheme, J, π, Nilsson assignments.

I. INTRODUCTION

The Brookhaven Medium Energy Intense Neutron facility[1] (MEIN) has made possible the production of several new neutron-rich nuclides,[2–5] particularly in the mass region above that of the fission products. The neutrons are produced by interaction of the 200-MeV proton beam (100 μA mean current) from the Linac injector of the Brookhaven Alternating Gradient Synchrotron with a water-cooled copper beam stop. The useful flux of these spallation neutrons with $E \geqslant 25$ MeV is $\approx 1.3 \times 10^{11}$ n cm^{-2} sec^{-1}. Sources of the new radionuclides were produced by $(n, 2pn)$ reactions and isolated with sufficient purity and in sufficient intensity to permit partial characterization of the decay properties despite the generally short half-lives of these nuclides. Previous studies concentrated on new even-even product nuclei whose predicted low β-decay energies indicated that their lifetimes would be sufficiently long to permit their isolation by radiochemical means and characterization by γ- and β-ray spectroscopy.

Extension of studies of this type into the heavy element region is of particular interest for several reasons: (1) broadening of nuclear structure systematics to provide comparisons with models which treat the collective and quasiparticle modes of nuclear excitation in heavy nuclei; (2) characterization of the nuclear mass surface for refinement of nuclidic mass models; (3) providing complementary energy level data by radioactivity measurements for nuclei which cannot be adequately studied by either neutron capture γ-ray or charged-particle reaction spectroscopy; (4) providing additional input data for a more reliable prediction of the nuclear properties of the postulated superheavy elements; and (5) supplying parameters needed for calculations of nucleosynthesis of heavy elements in high neutron flux environments by the s and r processes.

The availability of ^{244}Pu as target material and consideration of nuclear mass systematics and of probable $\log ft$ values indicated the feasibility of producing ^{242}U and ^{242}Np for study by means of ^{244}Pu$(n, 2pn)^{242}$U $\xrightarrow{\beta^-}$ ^{242}Np. This work led to the discovery and characterization of these two nuclei,[6] which are the heaviest known isotopes of U and Np. New levels in ^{242}Pu were found in addition to those which had been deduced previously from nuclear reaction studies.[7–9] Information on the levels of ^{242}Np was obtained for the first time. Level schemes were constructed and discussed within the framework of the collective model and Nilsson orbital systematics.

II. EXPERIMENTAL METHODS

In order to produce and isolate sources of ^{242}U and ^{242}Np from neutron-irradiated ^{244}Pu targets it was necessary to overcome a number of difficulties. The rare target material had to be recovered and recycled, and it had to be irradiated in a chemical form from which uranium could be separated rapidly. Extremely thorough chemical purifications were required because the cross section for the $(n, 2pn)$ reaction is very small compared to the production cross sections of fission products, Np isotopes, and other Pu isotopes which were produced during the irradiations. Furthermore, 14.1-h ^{240}U and its 7.2-min ^{240}Np^m daughter were constantly produced from the ^{244}Pu target by its α decay; their intense radiations were expected to interfere seriously with measurements of the much smaller amounts of ^{242}U and ^{242}Np that were produced. All of these problems were solved, or at least minimized, by the methods and procedures described below.

A. Targets and irradiations

The Pu target material (100–200 mg, 98.6% ^{244}Pu) in the +4 oxidation state was adsorbed from 7.5N nitric acid onto DOWEX MP-1 anion exchange resin (2–3 g) contained in a quartz cartridge. Immediately before the irradiations the ^{240}U which had grown in by α decay from ^{244}Pu was eluted off the resin with more nitric acid. The cartridge was sealed in Mylar and irradiated for 30 min in the MEIN facility at an effective flux of 1×10^{11} n cm^{-2} sec^{-1} (30–160 MeV). Transfer to and from the irradiation position was by means of a pneumatically operated rabbit.

B. Chemical separations and source preparation

Immediately after the irradiation uranium was chemically separated from plutonium, from fission products, and from other contaminants.[10] The principal steps were (1) elution of the U from the cartridge with 7.5N nitric acid leaving target Pu behind as Pu^{+4}, (2) adsorption of U onto a column of DOWEX MP-1 anion exchanger and washing with 10N nitric acid to remove most fission products, (3) elution of the U with 3N HNO_3, (4) solvent extraction of the U with ethyl acetate adsorbed on a polystyrene divinyl benzene chromatographic column, (5) adsorption onto another MP-1 column and washing with 6N HCl, and (6) elution of the U with water. Additional operations involved various evaporations to effect volume reductions, valence changes (e.g., Pu^{+6} to Pu^{+4}), and volatilizations of Ru and Tc contaminants. A special precipitation was needed to remove Te completely. The equilibrium source of U-Np was counted either as a solution in a flat glass vial or it was dried onto a glass disk. This procedure was performed in ~50 min; chemical yields, based on 0.2-0.5 mg added U carrier, were ~50%.

When it was desirable to study the ^{242}Np separated from its ^{242}U parent, additional chemical operations were needed. The uranium was adsorbed onto another MP-1 column from 2M acetic acid. After allowing an appropriate time for the ^{242}Np daughter to grow in, it was "milked" by elution with more 2M acetic acid. The parent ^{242}U is adsorbed very strongly by the MP-1 resin, so that its radiations can be emphasized strongly over those of the ^{242}Np by continuously flushing the column with acetic acid (see below).

Each quartz cartridge with the ^{244}Pu adsorbed as Pu^{+4} on the resin was usually used for two irradiations. Then the Pu was removed by reduction to Pu^{+3} with 5% hydroxylamine-hydrochloride and elution with 0.35N nitric acid.

C. γ-ray measurements

Several calibrated Ge(Li) detectors of large volume (≥ 50 cm^3) and high resolution [full-width at half-maximum (FWHM) $\approx$2.0 keV at 1332 keV] were used for the γ-ray spectroscopy. The low energy region (<100 keV) was also studied with a thin Be window Ge(Li) x-ray detector of ~0.8-keV resolution at 60 keV. The data were collected on magnetic tape by means of computerized 4096-channel analyzers. Energy and efficiency calibrations were made with National Bureau of Standards γ-ray sources. Data analysis was done by means of the SAMPO code[11] for γ-ray spectra and the CLSQ code[12] for decay curve fitting.

D. Beta-ray measurements

A plastic scintillator 2.5 cm thick attached to a photomultiplier tube was used as a detector for the β rays. Energy calibrations were made with sources[13] of ^{204}Tl, ^{210}Bi, and ^{144}Ce-Pr. The absolute β-disintegration rate of the ^{144}Pr (end point of 2997 keV) was determined by counting a very thin source (<10 $\mu g/cm^2$) in a 4π gas proportional counter; it was then used to calibrate the efficiency of the plastic scintillator. In addition the end point of the $^{240}Np^m$ β spectrum[14] was used as an internal standard, both for energy calibration and for correcting systematic errors due to source thickness and pulse pileup in the high counting geometry required.

III. EXPERIMENTAL RESULTS

Most of the γ-ray measurements were done with sources in which the ^{242}U and its ^{242}Np daughter were in secular equilibrium. The most intense peaks were from ^{237}U, ^{239}U, and ^{240}U-$^{240}Np^m$. In the early runs (with 10–30 mg Pu targets) two weak peaks were observed, at 735.9 keV and 780.4 KeV, which could not be attributed to known uranium isotopes or their neptunium daughters. Transitions of these energies were seen previously in studies[9] of $^{241}Pu(n,\gamma)^{242}Pu$; furthermore, a level at 781 keV had been observed in $^{242}Pu(d,d')$ experiments[8] which was assigned to $I^{\pi}=1^-$ in a $K=0$ octupole vibrational band. Tentatively, it was assumed that β decay of ^{242}Np, a short-lived daughter of ^{242}U, was feeding this same level. It was also noted that the 44.5-keV energy difference

(780.4–735.9) corresponds to the well established 2^+-0^+ transition in the $K=0$ ground state rotational band of ^{242}Pu.

The new γ rays decayed with a half-life of about 17 min and it was presumed that this is characteristic of the ^{242}U parent. The intensities of these peaks relative to peaks from ^{237}U and ^{239}U remained constant from run to run; thus the new lines were not caused by impurities and indeed they must be related to an isotope of U, or to its short-lived Np daughter. Chemical separations of Np from U soon confirmed that the 735.9- and 780.4-keV γ rays followed decay of the daughter and that the Np half-life was about 2 min. Figure 1 shows the decay of Np fractions milked periodically from the U parent. Subsequent experiments, with more target material, revealed 14 γ rays from ^{242}U and 39 γ rays from ^{242}Np. Initial counting rates were usually about 100 min^{-1} in the 735.9-keV peak.

A. γ-ray spectra and half-lives

The highest γ-ray intensities were obtained from equilibrium sources, but the "cleanest" spectra, those with fewer and smaller extraneous peaks, were obtained from the continuously milked sources. Figure 2 shows the γ spectrum from a U source on an ion exchange column placed next to a Ge(Li) detector while the Np daughter activities were being flushed out with acetic acid. The peaks from 23.5-min ^{239}U are identified and those from 6.75-d ^{237}U are indicated by an asterisk. All

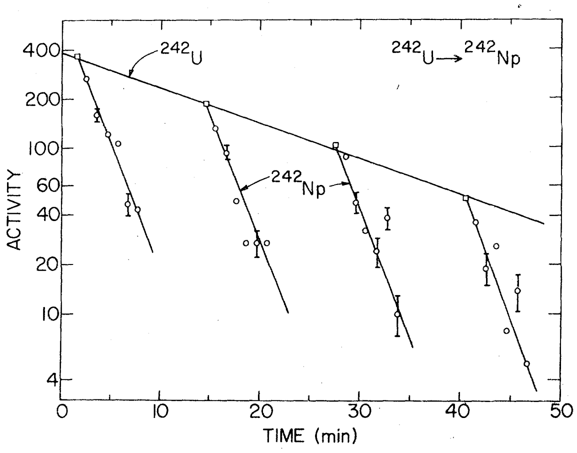

FIG. 1. Decay of ^{242}Np fractions milked periodically from ^{242}U parent. The measured activities correspond to sum of 735.9- and 780.4-keV γ rays; they are indicated by circles and typical error bars are shown. Extrapolations to the times of separation from U are shown by squares. The genetic relationship is demonstrated and within experimental errors the half-lives agree with the other measurements.

peaks attributed to ^{242}U are labeled with the appropriate energy in keV (620.6 is in parentheses because this peak is from decay of residual ^{242}Np). Other peaks[14] which are not labeled are from ^{240}U-Np. All 14 of the ^{242}U γ rays which have been established are listed in Table I. The intensities were derived from various sources measured with calibrated detectors. Figures 3(a) and 3(b) show the γ spectrum (up to 2 MeV) from Np which was being milked continuously from U on a column (behind a Pb shield) and flowed through a glass cell placed next to a Ge(Li) detector. The portion of the spectrum above 2 MeV [bottom panel in Fig. 3(b)] was obtained from measurements with equilibrium sources but the data are normalized to those from the milked source. Unlabeled peaks are from 7.2-min $^{240}Np^m$. The ^{242}Np results from all runs are summarized in Table II, where 39 γ rays are listed. Intensities of the parent and daughter γ rays shown in the tables were related by comparison with the 735.9-keV line and by making proper correction for the genetic relationship. In some cases small corrections were applied for coincidence summing; in other cases corrections were made for γ rays,[14,15] from $^{240}Np^m$ or ^{239}U, whose energies almost coincided with those from ^{242}U-Np. The uncertainties in γ energy are 0.1–0.3 keV in most cases while the errors in relative intensity vary between a few percent and 50%.

The more intense γ rays were used for half-life determinations of the parent and daughter. By combining data from several of the most intense runs we have by least square analyses[12] $T_{1/2}=16.8\pm0.5$ min for ^{242}U and $T_{1/2}=2.2\pm0.2$ min for ^{242}Np.

B. β-ray spectroscopy and Q_{β^-}

The β-ray measurements were made in order to determine Q_{β^-} for decay of the ^{242}Np and to establish a basis for estimating absolute γ-ray abundances. Since the β-ray intensity of $^{240}Np^m$ was very high and its spectrum[14] extends to 2.18 MeV it was possible to observe a pure β spectrum of ^{242}Np only above this energy. We postulated that a large fraction of the ^{242}Np β-decay strength would feed the ground state of ^{242}Pu and/or its low-lying 2^+ level, in a manner analogous to that observed in $^{240}Np^m$ decay[14] (52%). Theoretical predictions[16–18] of the β-decay energy of ^{242}Np range up to 3.0 MeV.

Figure 4 shows the higher energy portions of two β-ray spectra from a thin uranium source. The upper curve was obtained 1.0–1.5 h after the irradiation and the lower curve (normalized) was obtained later when the ^{242}U-Np had decayed away completely. Multiscaling of the β spectrum above 2.2 MeV resulted in a single component decay curve which exhibited the 17-min half-life char-

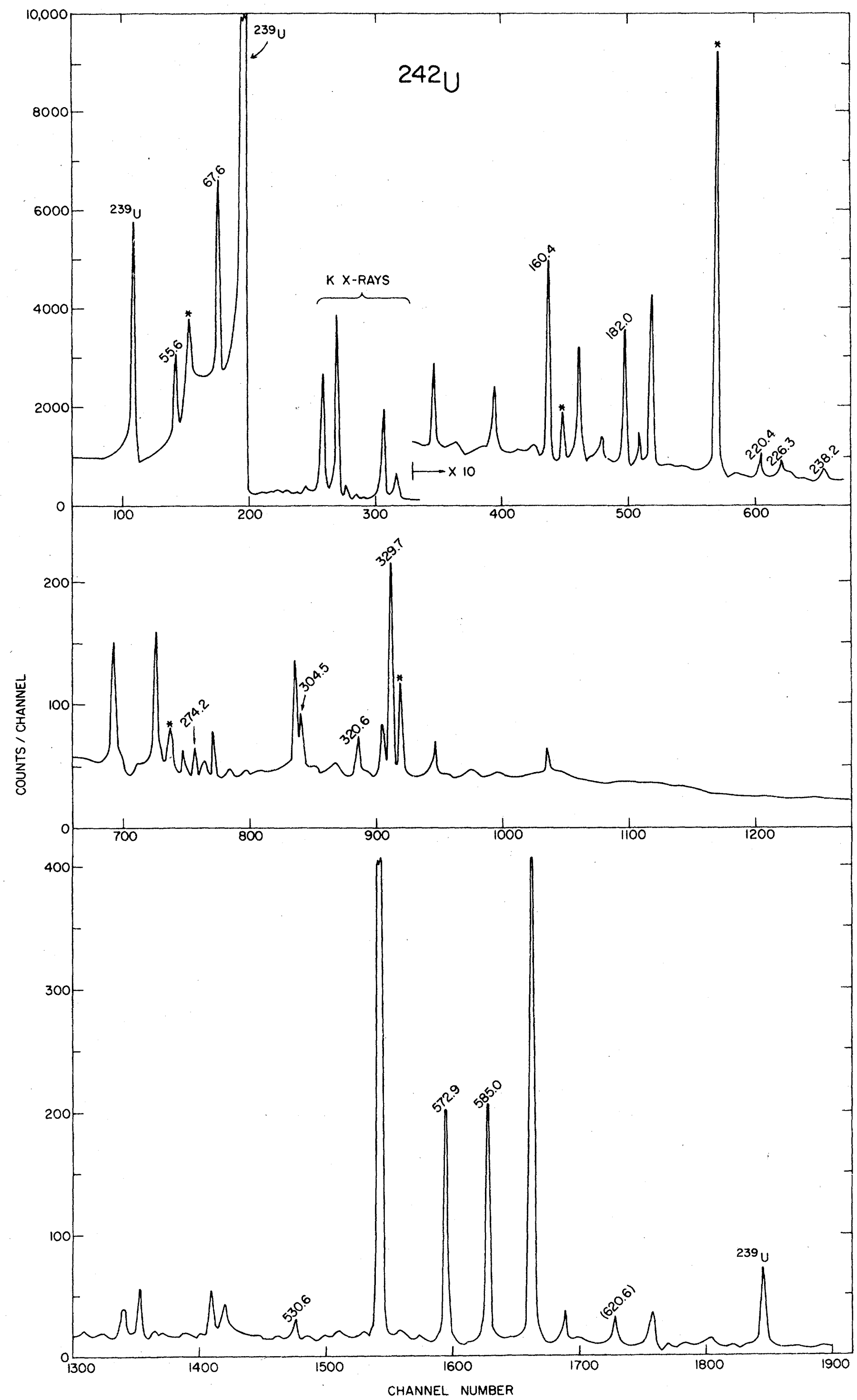

FIG. 2. γ-ray spectrum of a ^{242}U source from which Np daughter activity was being removed continuously. Energies in keV are given for each ^{242}U peak (the one at 620.6 keV is from residual ^{242}Np). The ^{239}U peaks are identified and those from ^{237}U are indicated by asterisks. The other peaks are from ^{240}U-$^{240}Np^m$.

TABLE I. γ-ray energies E_γ, in keV, and intensities I_γ from decay of 16.8-min ^{242}U. I_γ is relative to 100 for the 735.9-keV γ ray of the daughter ^{242}Np.

E_γ	I_γ	E_γ	I_γ
55.58 ± 0.06	75 ± 3	274.2 ± 0.2	2.2 ± 0.8
67.60 ± 0.05	184 ± 5	304.5 ± 0.2	6.8 ± 1.5
160.4 ± 0.1	15 ± 4	320.6 ± 0.1	4 ± 1
182.0 ± 0.1	14 ± 1	329.7 ± 0.1	15 ± 1
220.4 ± 0.3	3 ± 1	530.6 ± 0.2	4 ± 2
226.3 ± 0.1	2 ± 1	572.9 ± 0.1	36 ± 2
238.2 ± 0.1	4 ± 2	585.0 ± 0.1	37 ± 2

acteristic of the ^{242}U-Np pair in secular equilibrium. The end point energy of the ^{242}Np β rays was measured to be 2.7 ± 0.2 MeV. Assuming that there is appreciable decay directly to the 0^+ ground state, this value is also Q_{β^-}. If these higher energy β rays are predominantly from decay to the 2^+ level in ^{242}Pu, Q_{β^-} will be higher by 44.5 keV, but this difference is much smaller than the 200-keV uncertainty in the measurement. Thus, we conclude that $Q_{\beta^-} = 2.7 \pm 0.2$ MeV for ^{242}Np. Observation of the lower energy β spectrum of ^{242}U was not possible because it was masked by the

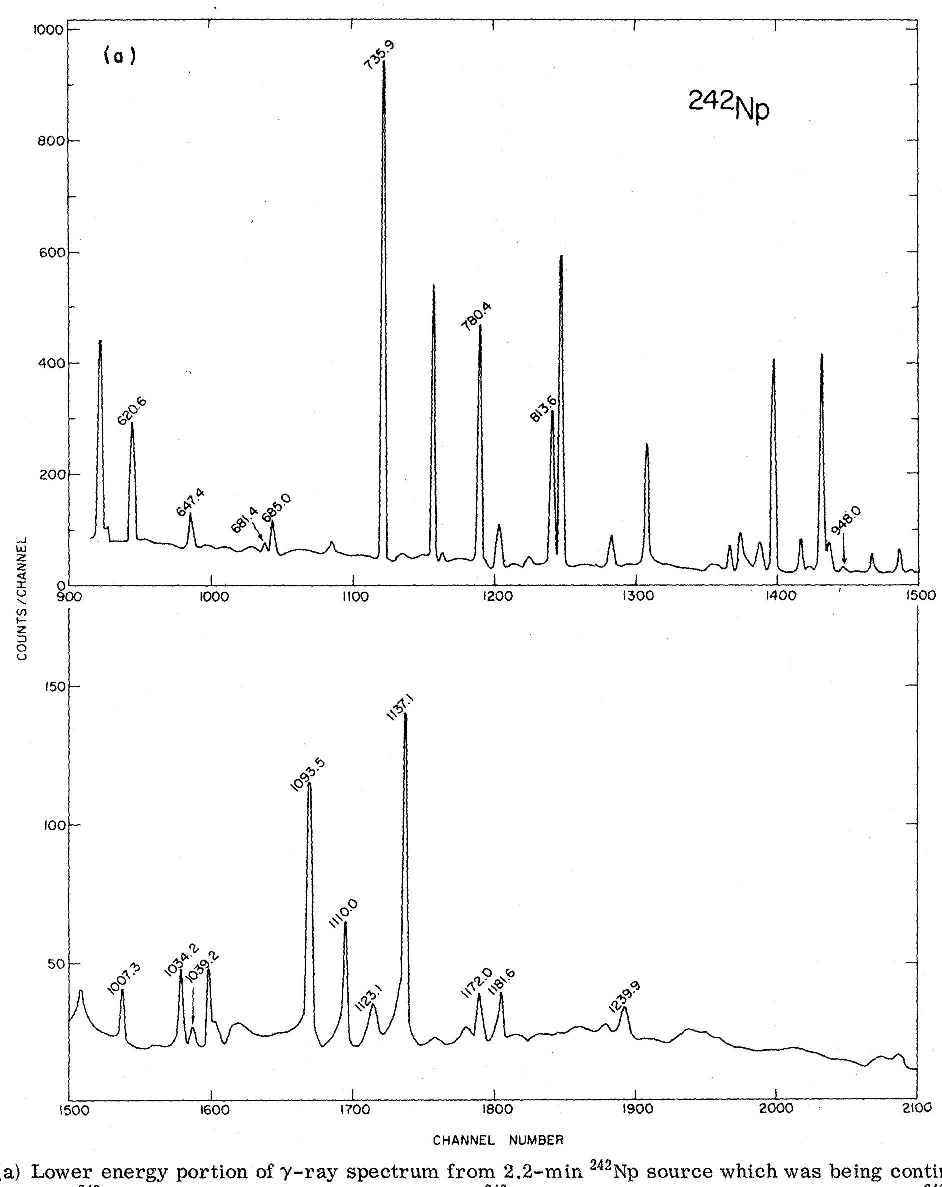

FIG. 3. (a) Lower energy portion of γ-ray spectrum from 2.2-min ^{242}Np source which was being continuously milked from its 16.8-min ^{242}U parent. Energies are given for the ^{242}Np peaks while the other peaks are from ^{240}Np^m. (b) Higher energy portion of γ-ray spectrum from 2.2-min ^{242}Np which was being continuously milked from its 16.8-min ^{242}U parent (upper two panels). The lowest panel was obtained from sources of ^{242}Np in secular equilibrium with ^{242}U but the activities are normalized to those of the upper panels. Energies are given in keV for each ^{242}Np peak.

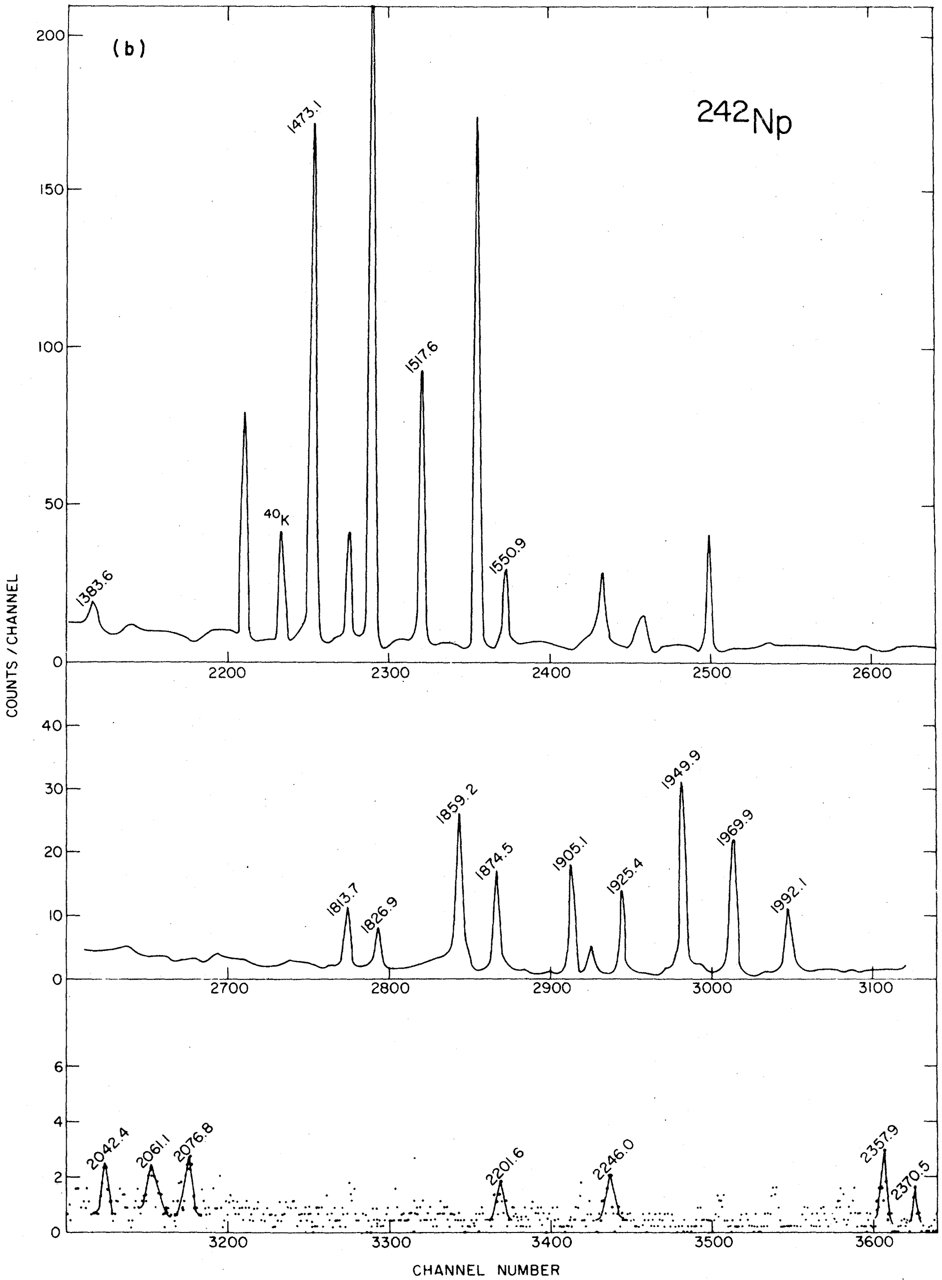

FIG. 3. (*Continued*).

much more intense radiations from ^{239}U and the ^{240}U-Np chain. β-γ coincidence measurements were not attempted because of insufficient activity levels.

Additional β-ray measurements, in combination with γ-ray counting of the same ^{242}U-Np source, were performed in order to estimate the fraction of ^{242}Np decays feeding the 0^+ and 2^+ states in ^{242}Pu. The total absolute intensity of the 2.7-MeV β disintegrations was derived from the portion of the spectrum above 2.18 MeV by comparison with the analogous portion of the ^{144}Pr β spectrum (E_{β^-} = 2.997 MeV) in the source of ^{144}Ce-Pr which was calibrated absolutely by 4π β counting. The absolute intensity of all the ^{242}Np β rays feeding excited levels in ^{242}Pu, at 780.4 keV and higher, was determined indirectly from the measured intensity of the 735.9-keV γ ray and the relative intensities

TABLE II. γ-ray energies E_γ, in keV, and intensities I_γ from decay of 2.2-min ^{242}Np. I_γ relative to 100 for the 735.9-keV γ ray.

E_γ	I_γ	E_γ	I_γ
620.6 ±0.1	18 ±2	1517.6±0.1	24 ±1
647.4 ±0.3	5.5±0.5	1550.9±0.1	7 ±1
681.4 ±0.4	2.9±1.0	1813.7±0.2	3.5±0.5
685.0 ±0.1	7 ±1	1826.9±0.3	2.3±0.5
735.93±0.07	100	1859.2±0.3	11.0±0.5
780.44±0.05	53 ±1	1874.5±0.3	5 ±1
813.6 ±0.1	24 ±2	1905.1±0.2	5.5±0.5
948.0 ±0.2	1.7±0.5	1925.4±0.2	4.5±0.5
1007.3 ±0.2	3.0±0.5	1949.9±0.2	14.8±0.5
1034.2 ±0.2	5.5±1.0	1969.9±0.2	10.5±0.5
1039.2 ±0.3	2.2±0.5	1984.5±0.5	1.0±0.2
1093.5 ±0.1	23 ±2	1992.1±0.3	4.0±0.2
1110.0 ±0.2	7 ±1	2042.4±0.7	0.8±0.2
1123.1 ±0.2	5 ±1	2061.1±1.0	0.6±0.2
1137.1 ±0.1	25 ±1	2076.8±0.5	1.3±0.3
1172.0 ±0.3	2.9±0.6	2201.6±0.5	1.2±0.3
1181.6 ±0.2	3.0±0.5	2246.0±0.5	0.9±0.3
1239.9 ±0.1	4.9±0.5	2357.9±0.5	1.0±0.5
1383.6 ±0.4	2.5±1.0	2370.5±0.5	1.0±0.5
1473.1 ±0.1	45 ±1		

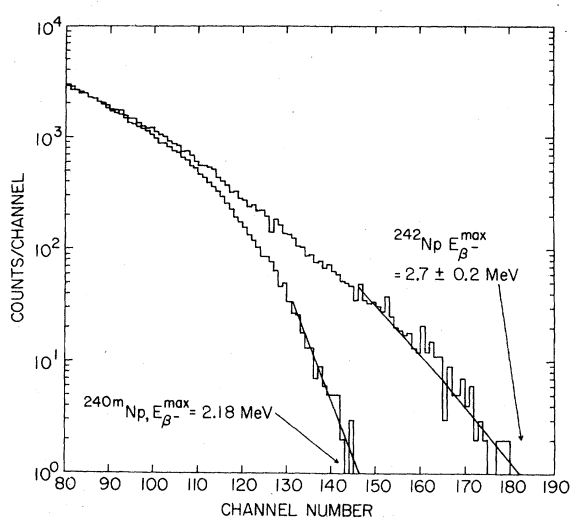

FIG. 4. Upper portions of the β-ray spectra of ^{240}Np^m and ^{242}Np. The end point of the latter is 2.7 ±0.2 MeV and is based on comparison with 2.18 MeV for ^{240}Np^m and with other standards.

of all the γ transitions (see decay scheme below). Thus, the fraction of all ^{242}Np β decay to the 0^+ ground state and the 44.5-keV 2^+ level is 83 ± 5%. This value is not sensitive to uncertainties in the decay scheme, nor do the β rays feeding the $I^\pi = 1^-$ level at 780.4 keV affect the analysis because their end point energy (1.92 MeV) falls below the energy interval that was used. The 83% value was then applied to calculate approximate absolute γ-ray abundances and the fractions of β-ray feeding into various levels (Figs. 5 and 6).

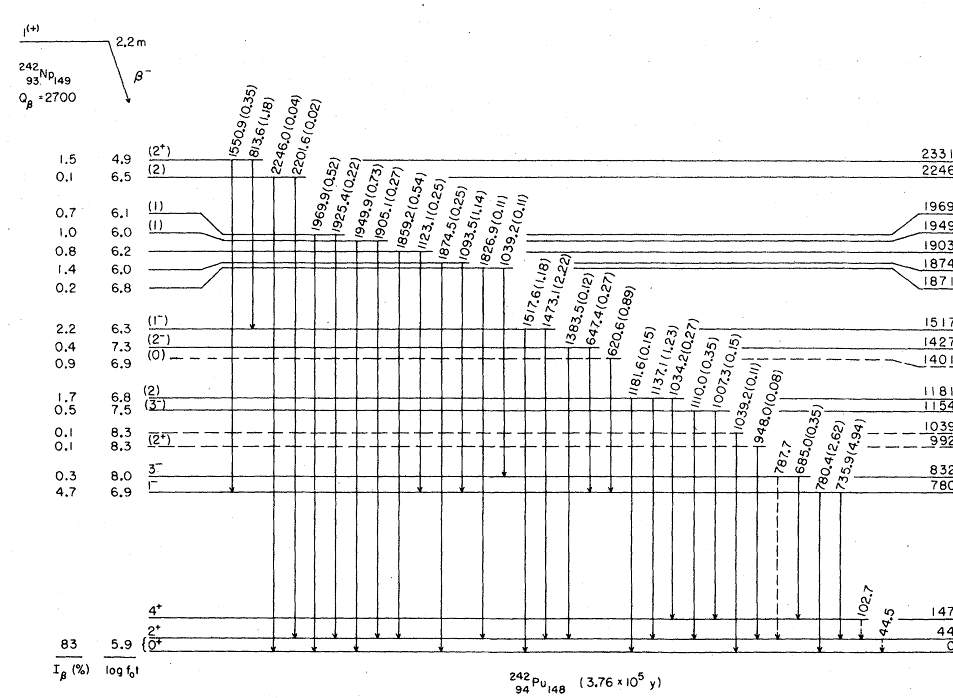

FIG. 5. Decay of ^{242}Np and proposed level scheme of ^{242}Pu. Figures in parentheses refer to γ-ray intensities per 100 ^{242}Np decays.

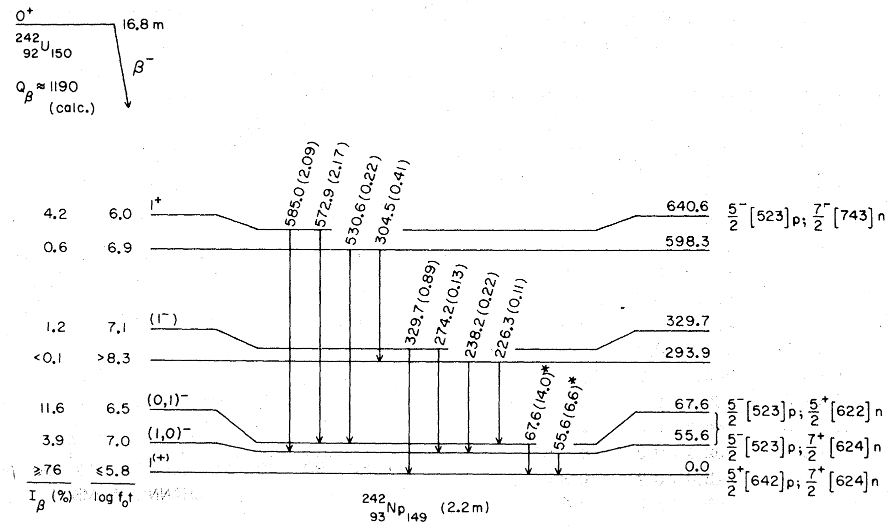

FIG. 6. Decay of ^{242}U and proposed level scheme of ^{242}Np. The figures given in parentheses refer to γ-ray intensities per 100 ^{242}U decays, except that for 55.6 and 67.6 keV (indicated by asterisks) the estimated transition intensities are given.

C. Effective cross sections for ^{244}Pu(n, $2pxn$) reactions

The effective cross sections σ_{eff} for production of the various uranium isotopes from the ^{244}Pu target were determined from several of the runs. Neutron flux measurements were made by assay of ^{24}Na induced in small aluminum monitor foils; σ_{eff} for ^{27}Al(n, α)^{24}Na was taken as 40 mb.[1] Absolute disintegration rates were determined for ^{237}U, ^{239}U, ^{240}U, and ^{242}U from the intensities of the strongest γ rays and their known abundances. Corrections were made for detector efficiencies and chemical yields. For ^{240}U a large additional correction (~50%) was made for the amount formed from the α decay of ^{244}Pu. A similar correction (10–15%) was needed for growth of ^{237}U from ^{241}Pu (present as 0.074 atom % of the target). The results are shown in Table III and are based on 6 runs.

IV. DISCUSSION AND CONCLUSIONS

Because of insufficient source intensity and relatively short half-lives, it was not practical to perform γ-γ and β-γ coincidence measurements. Thus, the proposed level schemes were constructed with the help of energy sums and differences, and intensity balances. Several previously known levels[9] in ^{242}Pu, and analogies with the nuclear systematics of neighboring nuclei provided a framework for characterizing some of the levels.

TABLE III. Effective cross sections σ_{eff}, in mb, for (n, $2pn$) and (n, αxn) reactions from ^{244}Pu target.

Product	Reaction	σ_{eff} (mb)
16.8-min ^{242}U	(n, $2pn$)	0.6 ± 0.3
14.1-h ^{240}U	(n, αn)	2.5 ± 1.2
23.5-min ^{239}U	(n, $\alpha 2n$)	1.8 ± 0.6
6.75-d ^{237}U	(n, $\alpha 4n$)	1.7 ± 0.7

Decay of 2.2-min ^{242}Np to levels of ^{242}Pu is shown in Fig. 5. The 44.5-keV transition, which is highly converted, was not seen directly, but six pairs of γ rays were observed which differed in energy by 44.5 keV. In most cases the ratios of reduced transition probabilities for these pairs agreed with theoretical predictions (Table IV). The 4^+ level at 147.2 keV is fed by three γ rays, but the 102.7-keV transition to the 2^+ level was not seen because it is strongly converted and a weak γ ray at this energy is masked by the intense K x rays. Higher levels of the ground state rotational band were not observed. The $K = 0$ octupole vibrational band is represented by the 780.4-keV 1^- level and the 832.2-keV 3^- level. These two and the 992.5-keV level also had been seen in the ^{242}Pu(d, d') studies.[8] In addition to all of these levels (up to 992.5 keV) the ^{241}Pu(n, γ) experiments[9] also indicated the 1039.2-keV level. The 787.7-keV line which is the crossover transition from 832.2 keV to 44.5 keV was not identified due to a strong interference from a γ ray at 789.6 keV in the decay of ^{240}Np^m. A dashed line was used to indicate a level whose position in the scheme is based on only a single γ-ray energy. The levels shown above 1039.2 keV were not found in previous work, and they were placed on the basis of intensity and energy sums. Estimates of β-ray feeding to the various levels were deduced from γ-ray intensity balances (to and from each level). Thus, logft values could be

TABLE IV. Comparisons of ratios of reduced transition probabilities in the ^{242}Np decay to predictions of the Alaga rule (Ref. 19).

Level (keV)	E_{γ_1} / E_{γ_2}	I_{γ_1} / I_{γ_2}	Exp. $B(E_{\gamma_1})/B(E_{\gamma_2})$ $L=1$	$L=2$	Theor. $B(E_{\gamma_1})/B(E_{\gamma_2})$ $L=1$	$L=2$	Assignment $K_iI_i^\pi \to K_fI_f^\pi$
780.4	735.9 780.4	100 53	2.25	2.53	2.0	···	$01^- \to 02^+$ $01^- \to 00^+$
1517.6	1473.1 1517.6	45 24	2.06	2.18	2.0	···	$01^- \to 02^+$ $01^- \to 00^+$
1949.8	1905.1 1949.9	5.5 14.8	0.40	0.42	0.50	···	$11 \to 02^+$ $11 \to 00^+$
1969.9	1925.4 1969.9	4.5 10.5	0.46	0.48	0.50	···	$11 \to 02^+$ $11 \to 00^+$
2246.0	2201.6 2246.0	1.2 0.9	1.43	1.47	···	1.43	$22 \to 02^+$ $22 \to 00^+$

calculated and in some cases spin-parity assignments could be made. Most of the γ rays could be placed in the decay scheme and they account for nearly all of the intensity. However, 11 of the weak lines were not placed.

The decay pattern of ^{242}Np to levels in ^{242}Pu exhibits a number of features in common with the decay of ^{240}Np^m to levels[14] of ^{240}Pu. The large β-ray branch from ^{242}Np to the ground 0^+ and/or first excited 2^+ level of ^{242}Pu is strong indication of a low spin for the ^{242}Np ground state. In addition, a significant β-ray branch to the 1^- octupole state in ^{242}Pu, analogous to the ^{240}Np^m decay, again suggests a low spin for ^{242}Np. Logft values for these β rays from ^{242}Np fall within the range for allowed-hindered or first forbidden transitions, as they do for ^{240}Np^m decays. The very weak β branch to the 3^- member of the octupole band of ^{242}Pu suggests that the ^{242}Np spin is ≤ 1. A similar analysis for ^{240}Np^m led previous investigators[14] to assign a spin-parity of 1^- to 7.2 min ^{240}Np^m formed by coupling the $\frac{5}{2}^-[523]_p$ and $\frac{5}{2}^+[622]_n$ Nilsson states. They assigned 5^+ to the 65-min ^{240}Np ground state formed from $\frac{5}{2}^+[642]_p$, $\frac{5}{2}^+[622]_n$, and its energy is just below that of the 1^- isomer. Thus, there is near degeneracy of the $\frac{5}{2}^+[642]$ and $\frac{5}{2}^-[523]$ proton states and a deformation $\epsilon \approx 0.2$ is indicated. The $N=149$ odd-mass isotones of ^{242}Np exhibit ground state spin-parities of $\frac{7}{2}^+$, suggesting occupation of the $\frac{7}{2}^+[624]_n$ Nilsson orbital ($\epsilon \approx 0.2$) by the odd neutron. The resultant coupling of these two states ($\frac{7}{2}^+[624]_n$ and $\frac{5}{2}^+[642]_p$) yields a spin-parity of 1^+ for the 2.2-min ^{242}Np. It is important to note that the present work cannot establish whether this state is the nuclide's ground state or an isomeric state. Whether a high spin isomer exists in ^{242}Np (analogous to the 65-min 5^+ state in ^{240}Np) remains to be established by future studies in which the Np is isolated directly from neutron irradiated ^{244}Pu.

Having assigned $I^\pi = 1^+$ to 2.2-min ^{242}Np, it is possible to discuss the levels of ^{242}Pu in more detail (Fig. 5). Below 850 keV of excitation energy ^{242}Pu exhibits a ground state rotational band ($\hbar^2/2\mathscr{I} = 7.41$ keV) and a $K^\pi = 0^-$ octupole band with 1^- band head located at 780.4 keV ($\hbar^2/2\mathscr{I} = 5.18$ keV). Logft values for β decay to the ground state band and to the octupole band are in the range expected for allowed and first forbidden transitions, respectively. The 5^- member of the octupole band (924.5 keV) is not observed in the present experiment, nor is the 0^+ state previously reported at 956 keV from (p,t) reaction studies.[7] The 2^+ state at 992.5 keV seen in the (d,d') experiments[8] is weakly fed (~0.1%, log$ft = 8.3$) from the ^{242}Np decay. Excited states above 1.1 MeV decay predominantly to either the levels of the ground state band or the octupole band. Logft values for these states in combination with reduced transition probabilities for γ rays de-exciting these levels to the 0^+ and 2^+ levels of the ground state band have been compared to predictions of the Alaga model[19] to establish limiting spin-parity assignments for these states. A number of these comparisons are shown in Table IV. The decay pattern of the 1181.6-keV level is puzzling. γ rays from this state to the 0^+, 2^+, and 4^+ levels of the ground state band indicate that this state is most probably $I=2$. The strong γ-ray branch to the 2^+ level over that to either 0^+ or 4^+ levels is, however, anomalously high and may indicate that the 1137.1-keV transition is, in fact, a composite of the 1181.6 → 44.5-keV decay and another moderately intense γ ray which may occur elsewhere in the decay scheme.

A particularly striking feature in the ^{242}Np decay is the highly favored β transition to the 2331.3-keV level of ^{242}Pu. The low logft (4.9) for this branch suggests that the 2331.3-keV level is of distinctly different character from the nearby levels which, at ≈2 MeV of excitation, are viewed as part of the ensemble of states derived from various statistically allowed combinations of single particle and/or collective excitations. The highly favored β decay to the 2331.3-keV level and the atypical decay pattern of this level to lower-lying states in ^{242}Pu point to a unique quasiparticle configuration for this state. From consideration of a number of two quasiparticle configurations which can be constructed out of the available Nilsson orbitals at this excitation energy, a 2^+ two-proton state $\frac{5}{2}^+[642]_p$, $\frac{9}{2}^+[624]_p$ is postulated as a likely assignment for the 2331.3-keV level. The low logft is consistent with the allowed, unhindered β decay: ^{242}Np $\frac{5}{2}^+[642]_p$, $\frac{7}{2}^+[624]_n$ $\rightarrow$ ^{242}Pu(2331.3 keV) $\frac{5}{2}^+[642]_p$, $\frac{9}{2}^+[624]_p$, in which no change occurs in the asymptotic quantum numbers of the nucleons involved in the β transition.

Additional insight can be obtained from the γ-ray de-excitation pattern of this 2331.3-keV state. Two transitions (1550.9 and 813.6 keV) are observed to lower-lying 1^- levels. On energetic grounds one would expect the 1550.9-keV transition to be considerably more intense than the 813.6-keV transition, but, in fact, the opposite is true. This is taken as an indication of the degree of dissimilarity between the quasiparticle nature of the 2331.3-keV state and the collective nature of the 1^- octupole state and at the same time the similarity between the 2331.3-keV state and the 1517.6-keV level, which by analogy with neighboring Pu isotopes has been assigned 1^-. The smaller logft (6.3) to the 1517.6-keV over that to the octupole band head level at 780.4 keV (logft = 6.9) is again suggestive of a quasiparticle structure for the 1517.6-keV level. We postulate a two-proton configuration $\frac{5}{2}^+[642]_p$, $\frac{5}{2}^-[523]_p$, for this level, based largely on the availability of Nilsson orbitals at this excitation energy, the unretarded β transition $\frac{7}{2}^+[624]_n \rightarrow \frac{5}{2}^-[523]_p$ from ^{242}Np, and the favored γ-ray transition from the 2331.3-keV state, which for one of the nucleons, again involves no change in asymptotic quantum numbers. In an analogous way the decay of the 1401.0-keV state by a single 620.6-keV γ-ray transition to the 1^- ($K=0$) octupole band head suggests a zero spin for the 1401.0-keV level. Logft arguments cannot establish the parity of this state.

A few comments on the character of the vibrational structure of ^{242}Pu seem appropriate. It is noteworthy that our present β-decay results do not indicate either well developed β-($K=0$) or γ-($K=2$) vibrational bands in ^{242}Pu. Population of the octupole vibration in ^{242}Pu as well as in other even-even actinide nuclei is, however, a dominant feature of the β decay. Why the negative parity collective vibration is well developed in ^{242}Pu in preference to either of the positive parity vibrations is a puzzle. The combination of a 1^+ spin-parity for ^{242}Np and the β-decay selection rule of $\Delta K = 0, 1$ makes the ^{242}Np decay an excellent place to look for the relative importance of the various vibrational modes of excitation. The known $I^\pi = 0^+$ "collective" level[7] at 956 keV was not observed in the β decay of ^{242}Np. Population of the 992.5-keV, $I^\pi = 2^+$, $K=0$ level is questionable because in our work this level is introduced only on the basis of the weak 948.0-keV γ ray which concomitantly sets a high logft of 8.3 to the 992.5-keV level. While Table IV indicates the presence of a high-lying $K = 2$ band head, its excitation energy clearly rules it out as a γ vibration. These features and the fact that the collective octupole band head is relatively strongly populated by a first forbidden β-ray transition in preference to allowed β decays to positive parity vibrations indicates that the collectivity of these vibrations is relatively weak. In this case matrix elements of β- and γ-ray transitions are mostly of quasiparticle origin, and therefore they are regulated by the usual asymptotic Nilsson selection rules, and these transitions are usually retarded.

Negative parity collective states (octupole vibrations) are known to be strongly collective in heavy nuclei [$B(E3)$ is large from Coulomb excitation] and therefore quasiparticle selection rules for β and γ transitions to these states (or from these states) are expected to be not so effective, and retardation of these transitions is smaller. Therefore we observe usual values for logft's. Additional evidence for this type of collective behavior is to be found in the pattern of the electron capture decay[20] of ^{238}Am to levels in ^{238}Pu. The ground state of ^{238}Am has been assigned as 1^+ and expect for ^{242}Np it is the only other actinide nuclide with this spin-parity. One again observes very heavy feeding of the odd-parity collective states by first forbidden transitions in preference to allowed decays to the even parity collective states.

It is important to note that following β decay to other low spin ($I \leq 2$) levels no γ transitions to the $I^\pi = 0^+$ ground state were observed. If their spin is $I=2$, it is more natural to expect that they have negative parity, $I^\pi = 2^-$. It is also interesting to compare groups of 2 levels in ^{242}Pu: 1401.0 ($I=0$) and 1427.8 keV ($I=2$) and in ^{240}Pu: 1410.8 ($I=0$) and 1438.5 keV ($I=2$). These levels in ^{240}Pu were considered as members of a rotational band with $K^\pi = 0^+$ and an octupole two phonon state of $K^\pi = 0^-$ origin. However, in ^{240}Pu some doubt about parity came from the (n,γ) data, which showed negative parity for these levels. Further support for negative parity comes from the β-decay data of ^{242}Np because in ^{242}Pu the $I=2$, 1427.8 keV level γ decays only to 2^+, but not to the 0^+ ground state. Viewed as levels with $I^\pi=0^-$ and 2^-, they may be considered as members of a rotational band with $K^\pi=0^-$. The moment of inertia of this band $\mathscr{g}$ is significantly larger than $\mathscr{g}$ for the ground state rotational band ($\mathscr{g}_{K=0^-}/\mathscr{g}_{K=0^+}=1.67$). This is typical for two particle configurations because unpaired nucleons in orbitals with large j tend to increase sharply the effective moment of inertia. One of the lowest possible configurations for such a $K^\pi=0^-$ state is $\frac{7}{2}^-[743]_n$, $\frac{7}{2}^+[624]_n$, where both unpaired neutrons are in the orbitals with large j and therefore strongly involved in Coriolis coupling which determines the effective moment of inertia.

No previous information concerning the level structure of ^{242}Np is available and our experimental data are not sufficient to establish an unambiguous decay scheme for ^{242}U. However, some levels of ^{242}Np can be deduced on the basis of γ-ray energy loops and γ-ray intensities. A proposed decay scheme is shown in Fig. 6 where 10 of the 14 γ rays are accounted for.

β-ray and γ-ray intensities were related to those which were measured for the ^{242}Np daughter in secular equilibrium. The values in parentheses associated with each γ ray are estimated γ-ray intensities per 100 decays, except for the 67.6- and 55.6-keV γ rays, where transition intensities are given. The latter transitions are almost certainly $E1$, and appropriate corrections were made for internal conversion. Higher multipole orders are excluded because the transitions would be so highly converted that the γ rays would be too weak to be observed. The total absolute γ-ray abundances account for about 30% of the total decay, and thus there must be a strong β branch to the ground state or there are some highly converted unobserved transitions in the ^{242}U decay. The percent of β^- feeding to various levels and the logft values given in Fig. 6 were calculated on the basis of 76% to the ground state and an estimated Q_{β^-}-value of 1.19 MeV (see below).

Some of the major features of the level scheme can be interpreted from Nilsson orbital systematics. From the 0^+ ground state of ^{242}U, the large β branch to the ^{242}Np 1^+ ground state is indicative of both the favorable energetics and the allowed nature of the decay. The low-lying negative parity states at 55.6 and 67.6 keV, which decay by $E1$ transitions to the ^{242}Np ground state are interpreted as odd proton and odd neutron configurations which involve the next available proton orbital, $\frac{5}{2}^-[523]$, and either the same neutron configuration as the ground state or a neutron hole from the $\frac{5}{2}^+[622]$ orbital. Decay of the 640.6-keV level through the 55.6- and 67.6-keV levels rather than directly to the ground state indicates a large structural difference between the 640.6-keV level and the ground state. Logft arguments suggest positive parity for this level. Coupling a $\frac{7}{2}^-[743]$ neutron hole to the $\frac{5}{2}^-[523]$ proton, which is responsible for the 55.6- and 67.6-keV states, results in a 1^+ assignment for the 640.6-keV level. The 585.0- and 572.9-keV γ rays are then seen as allowed $E1$ transitions involving either $\frac{7}{2}^-[743]_n \to \frac{5}{2}^+[622]_n$ or $\frac{7}{2}^-[743]_n \to \frac{7}{2}^+[624]_n$ transformations. The absence of the 640.6-keV ground state γ ray points to a considerable degree of forbiddenness for the simultaneous transitions of both neutron and proton configurations from this state to those of the ground state.

It is instructive to compare the measured Q_{β^-} of ^{242}Np of 2.7 + 0.2 MeV with predictions from several currently available mass calculations,[16–18,21,22] which are summarized in Table V. Within the experimental error, agreement is good with most of the predicted values; the first three listed are low by 0.4–0.5 MeV. Since we do not have an experimental value for Q_{β^-} of ^{242}U it was estimated by taking an average of the predicted values (omitting the first three). This gives 1.19 MeV and it was used to compute logft values in the ^{242}U decay (Fig. 6). The calculated value of $Q_\beta=0.24$ MeV for ^{242}U shown in Table V is clearly much too low since γ rays up to 0.585 MeV were observed in the ^{242}U decay.

TABLE V. Comparison of experimental and predicted Q_{β^-} values for the ^{242}Np and ^{242}U decays.

	^{242}Np	^{242}U
Experiment	2.7 ± 0.2 MeV	a
Seeger and Howard [b]	2.20	1.00 MeV
Myers [b]	2.26	0.24
Groote, Hilf, Takahashi [b]	2.33	0.67
Jänecke and Eynon [b]	2.48	0.97
Monahan and Serduke [c]	2.78	0.98
Jänecke (Garvey-Kelson) [b]	2.78	1.34
Comay and Kelson [b]	2.79	1.59
Wapstra and Bos [d]	2.80	e
Bauer [b]	2.88	e
Viola, Swant, Graber [f]	2.94	0.88
Liran and Zeldes [b]	2.97	1.36

[a] Not measured; 1.19 MeV used in the decay scheme was obtained by averaging the last six predicted values.
[b] Reference 21.
[c] Reference 22.
[d] Reference 18.
[e] Not predicted.
[f] Reference 16.

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[1]S. Katcoff, J. B. Cumming, J. Godel, V. J. Buchanan, H. Susskind, and C. J. Hsu, Nucl. Instrum. Methods 129, 473 (1975).

[2]E-M. Franz, S. Katcoff, H. A. Smith, Jr., and T. E. Ward, Phys. Rev. C 12, 616 (1975).

[3]P. E. Haustein, E-M. Franz, S. Katcoff, N. A. Morcos, H. A. Smith, Jr., and T. E. Ward, Phys. Rev. C 14, 645 (1976).

[4]P. E. Haustein, E-M. Franz, R. F. Petry, and J. C. Hill, Phys. Rev. C 16, 1559 (1977).

[5]J. Gilat and S. Katcoff, J. Inorg. Nucl. Chem. 40, 369 (1978).

[6]A preliminary report was presented at the Washington, D. C. meeting of the American Physics Society, 1978 (unpublished); S. Katcoff, E-M. Franz, P. E. Haustein, R. L. Klobuchar, and H-C. Hseuh, Bull. Am. Phys. Soc. 24, 614 (1978).

[7]J. V. Maher, J. R. Erskine, A. M. Friedman, R. H. Siemssen, and J. P. Schiffer, Phys. Rev. C 5, 1380 (1972).

[8]T. W. Elze and J. R. Huizenga, Nucl. Phys. A187, 545 (1972).

[9]Y. A. Ellis and R. L. Haese, Nucl. Data Sheets 21, 615 (1977).

[10]Most of the steps were derived by suitable modification of previous methods: J. E. Grindler, National Academy of Sciences—National Research Council, Nuclear Science Series Report No. NAS-NS-3050, 1962 (unpublished); K. Wolfsberg, Los Alamos Scientific Laboratory Report No. LA-1721, 1967 (unpublished); P. Van Den Windel, F. DeCorta, and J. Hoste, Anal. Chim. Acta 56, 241 (1971).

[11]J. T. Routti and S. G. Prussin, Nucl. Instrum. Methods 72, 125 (1969).

[12]J. B. Cumming, National Academy of Sciences—National Research Council, Nuclear Science Series Report No. NAS-NS-3107, 1962 (unpublished).

[13]β-ray and γ-ray energies and intensities were taken from the compilations in Nuclear Data Sheets.

[14]M. R. Schmorak, Nucl. Data Sheets 20, 165 (1977).

[15]M. R. Schmorak, Nucl. Data Sheets 21, 91 (1977); D. R. Mackenzie and R. D. Connor, Nucl. Phys. A108, 81 (1968).

[16]V. E. Viola, Jr., J. A. Swant, and J. Graber, At. Data Nucl. Data Tables 13, 35 (1974).

[17]A. H. Wapstra and K. Bos, At. Data Nucl. Data Tables 17, 474 (1976).

[18]A. H. Wapstra and K. Bos, At. Data Nucl. Data Tables 19, 175 (1977).

[19]T. Yamazaki, Nucl. Data A 1, 453 (1966).

[20]Y. A. Ellis, Nucl. Data Sheets 21, 549 (1977); I. Ahmad, R. K. Sjoblom, R. F. Barnes, F. Wagner ner, Jr., P. R. Fields, Nucl. Phys. A186, 620 (1972).

[21]Nine different mass formulas are discussed and compared in a special issue of At. Data Nucl. Data Tables 17, 411 (1976).

[22]J. E. Monahan and F. D. Serduke, Phys. Rev. C 17, 1196 (1977).