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Author's Note: This is a preliminary version of a review of the isotopes of thorium, protactinium and uranium which will later form part of a comprehensive treatment of nuclear data for the entire heavy element group of elements.

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8.1 GENERAL CONSIDERATIONS

This chapter consists of a systematic and detailed review of the radioactive properties of the isotopes of thorium, protactinium, and uranium. In the previous two chapters the presence of many of these isotopes in the heavy element decay series both natural and synthetic has been mentioned, but a complete discussion of their decay schemes has been reserved for this chapter. Appropriate cross-references to the two previous chapters are made in the following material.

It may be useful first to speak of some general features of these decay schemes which may make it easier to understand what otherwise might be a confusing compilation of data. Since general features are more clearly discernable in the nuclear disintegrations leading to an even-even daughter product, we start with comments about such cases. Figure 8.1 shows all the identified levels lying below 600 kev in the even-even nuclei on which data have been obtained by a study of the decay of isotopes of thorium, protactinium and uranium. The most common feature is a group of even parity states with the spin sequence 0+, 2+, 4+, etc. This series represents rotational excitation of the spheroidally deformed ground state nucleus. The spacing of the levels follows the rotational formula,

$$E_I = \frac{\hbar^2}{2\Im} I(I+1),$$

where $\Im$ is the effective moment of inertia. There is a steady decrease in the energy of the levels and hence a steady increase in the effective moment of inertia as the mass number increases. At mass number 232 the value of $\hbar^2/2\Im$ is about 7.5 kev and this value prevails nearly unchanged throughout the whole transuranium element group of even-even nuclei. Such nuclei are strongly stabilized in a deformed shape. In the case of the lightest nuclei here considered $\hbar^2/2\Im$ is about 15-20 kev. These nuclei may be just barely stabilized in a non-spherical shape.

The various features of the ground state rotational band, which are discussed fully in chapter 3, are observed in the cases under discussion here. For example, the pattern of gamma ray de-excitation is in every case a series of E2 cascade transitions with weak or zero competition from M1 radiation.

In the case of alpha decay of an even-even isotope the pattern of alpha decay is extremely regular. The most prominent alpha group is that leading to the ground state with an intensity of about 70 percent. About 30 percent of the transitions populate the first-excited 2+ state. Higher lying states are populated in very small intensity because alpha decay in even nuclei is subjected to a very strong and very regular dependence on alpha decay energy.

In several cases the decay of thorium, protactinium, or uranium isotopes by orbital electron capture or by beta decay produces an even-even daughter nuclei whose lowest-lying levels have been revealed by study of an alpha emitter. This knowledge assists greatly in the construction of the decay scheme of the beta-unstable nuclides. The decay of these nuclides tends to go more readily to higher lying states and, hence, to reveal a more complex spectrum of excited levels in the daughter nucleus.

Aside from the ground state rotational band the next most prominent feature of the level system is the negative parity band with a spin, parity sequence 1-, 3-, 5-. A great deal of evidence of the type discussed in chapter 3 suggests that this sequence represents rotational excitation superimposed on a vibrational mode of excitation, where the spin and parity assignments of the vibrational excitation is 1-. Furthermore it is believed that this vibration can be assigned to an octupole deformation in shape of the nucleus, a deformation which destroys the spheroidal symmetry of the ground state.

The spacing of the 1-, 3-, and 5- levels also follows an I(I+1) rotational law. The pattern of de-excitation consists of a pair of E1 transitions from each level of spin I to the levels of spin I-1 and I+1 in the even-parity ground state rotational sequence of states. The energy of the 1- states varies widely from nucleus to nucleus for reasons which are not well understood.

The unified model of the nucleus suggests that other types of collective vibrational motion might contribute to the levels of excitation in the even nuclei. Particularly in the cases of Th^{228}, Th^{230}, and Th^{232} evidence has been found for the occurrence of such levels somewhat above the energy region included in figure 8.1. There are two types of quadrupole deformation which have been identified in these thorium nuclei. These are the so-called beta and gamma vibrations. These vibrational types are

discussed fully in chapter 3 and their specific characteristics within the thorium nuclei are discussed in the following sections.

When we consider the decay of thorium, protactinium, and uranium isotopes to daughter isotopes containing an odd number of neutrons or protons, we can at present discern very little order in the system of levels. The interpretation of the observed levels in this particular mass region is much less clear than in the mass number range spanned by the transuranium elements. The nuclei of the transuranium element nuclides are firmly stabilized in a spheroidal shape. In such nuclei we can often clearly associate the observed energy levels with the theoretical single-particle wave functions calculated by NILSSON. Superimposed on these single-particle states of intrinsic motion we are frequently able to identify bands of rotational levels. However, the odd-mass nuclei formed by the decay of thorium, protactinium, and uranium isotopes fall in a transition region between the strongly deformed nuclei of the so-called strong-coupling region and the spherically symmetric nuclei of the simple shell model. The Nilsson wave functions are not so easy to identify with observed levels and the theoretical treatment of the expected level systems is more complex.

The level system of the odd-odd nuclei is even further from a theoretical interpretation because in general the treatment of the level system is such nuclei must be based on a clear knowledge of the levels in neighboring odd mass nuclei.

It is perhaps worth calling attention to the great complexity of the alpha spectra of the odd nuclei or odd-odd nuclei in this group of nuclides. Thorium 227 has a very complex spectrum consisting of 15 distinct groups. The alpha decay of Pa^{228} is even more complex since more than 25 alpha groups have been found. Since there exists no satisfactory description of the complex level systems of the daughter nuclei the radiations of such alpha emitters must be presented in the sections which follow without interpretive comment.

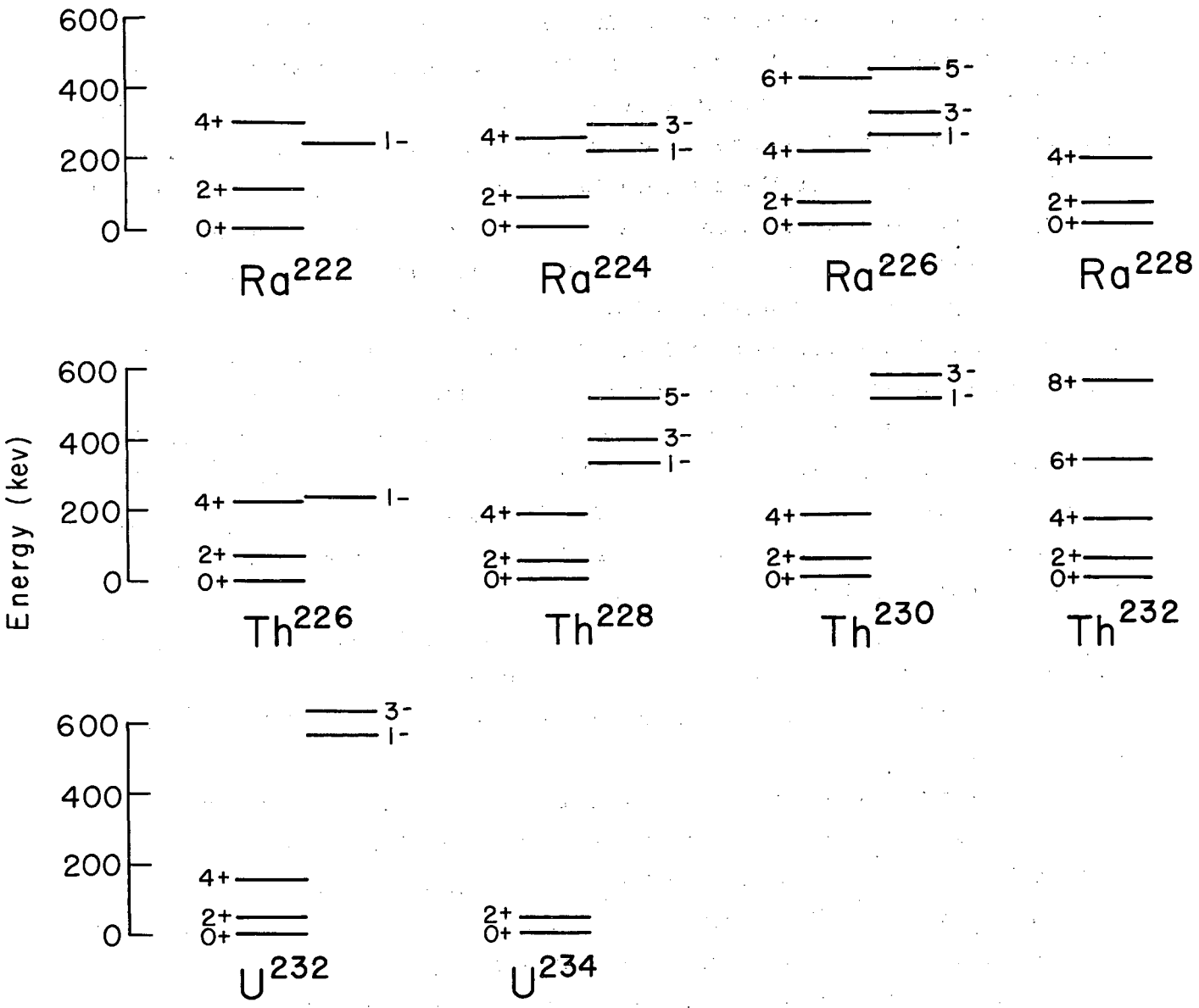

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Figure 8.1. System of levels up to 600 kev of excitation observed in nuclei produced by the decay of isotopes of thorium, protactinium or uranium. Only observed levels are shown. In the case of Th^{232} the level scheme was obtained from coulombic excitation experiments. This figure is meant to show the great regularity in the observed levels, all of which are interpreted as rotational excitation of the 0+ ground state and of a 1- vibrational state.

8.2 THE ISOTOPES OF THORIUM

8.2.1 Thorium-223. This isotope is a member of the U^{227} collateral series (See Section 7.2.8) and the only information available on its properties is that it emits alpha particles with an energy of 7.55 ± 0.1 Mev. The half-life, estimated from systematic trends in alpha decay half lives, is ~0.1 second.

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1. W. W. Meinke, A. Ghiorso, and G. T. Seaborg, Phys. Rev. 85, 429 (1952).

8.2.2 Thorium-224. This isotope is a member of the U^{228} collateral series (See Section 7.2.7). All that is reported about Th^{224} is that it emits alpha particles of 7.13 ± 0.02 Mev energy with a half-life estimated at 1 second from the systematic trends in alpha decay half-lives.

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1. W. W. Meinke, A. Ghiorso, and G. T. Seaborg, Phys. Rev. 81, 782 (1951).

8.2.3 Thorium-225. This isotope is a member of the U^{229} collateral series (See Section 7.2.6). It decays with a half-life of 8.0 ± 0.5 minutes. Ten percent of the disintegrations go by orbital electron capture to produce Ac^{225} and ninety percent go by the emission of 6.57 ± 0.03 Mev alpha particles to produce Ra^{221}.

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1. W. W. Meinke, A. Ghiorso, and G. T. Seaborg, Phys. Rev. 81, 782 (1951).

8.2.4 Thorium-226. This isotope is a member of the Pa^{230} collateral series[1] (See Section 7.2.1). It is an alpha emitter with a half-life of 30.9 minutes. The alpha spectrum as measured by ASARO AND PERLMAN[2] consists of four groups:

6.330 Mev	(79 percent)
6.220 Mev	(19 percent)
6.095 Mev	(1.7 percent)
6.029 Mev	(0.6 percent)

Examination of the gamma ray spectrum with a scintillation crystal spectrometer[2]

showed gamma rays of energy 112, 130, 197, and 242 kev belonging to Th^{226}. Other gamma radiations present in the sample were due to short-lived daughter activities. Gamma-gamma coincidence studies showed that the 112 kev gamma ray was in coincidence with the 130 and the 197 kev gamma rays but not with the 242 kev radiation.

All the alpha particle and gamma ray data on energies and abundances fit neatly the decay scheme shown in Fig. 8.2. The prominent 112 kev radiation was characterized as an electric dipole transition by its conversion coefficient,[2] and its high conversion in the L_{II}, L_{III}, M_{II}, and M_{III} shells,[4] and its angular correlation with the alpha particles leading to the state. SMITH, ASARO, AND HOLLANDER[4] report a more precise energy value of 111.1 ± 0.3 kev.

STROMINGER[5] performed alpha-gamma coincidence experiments which established that the 112, 197, and 242 kev gamma rays were emitted within a period of less than 1.4×10^{-9} seconds.

The 1- assignment of the 242 kev state was made definite by the alpha-gamma angular correlation experiments of STEPHENS, ASARO, AND PERLMAN[3] who found a clear correlation of the type predicted for the sequence:
$0^+ \xrightarrow{\alpha} 1^- \xrightarrow{\gamma} 0^+$.

All the levels seen in the decay of Th^{226} are clearly assignable to collective modes of motion of the nucleus. The 0+, 2+, 4+ sequence and the 1- state seen in this instance are of similar origin to comparable states seen in the decay of Th^{228}, Th^{230}, U^{230} and other even-even nuclei in this mass region.

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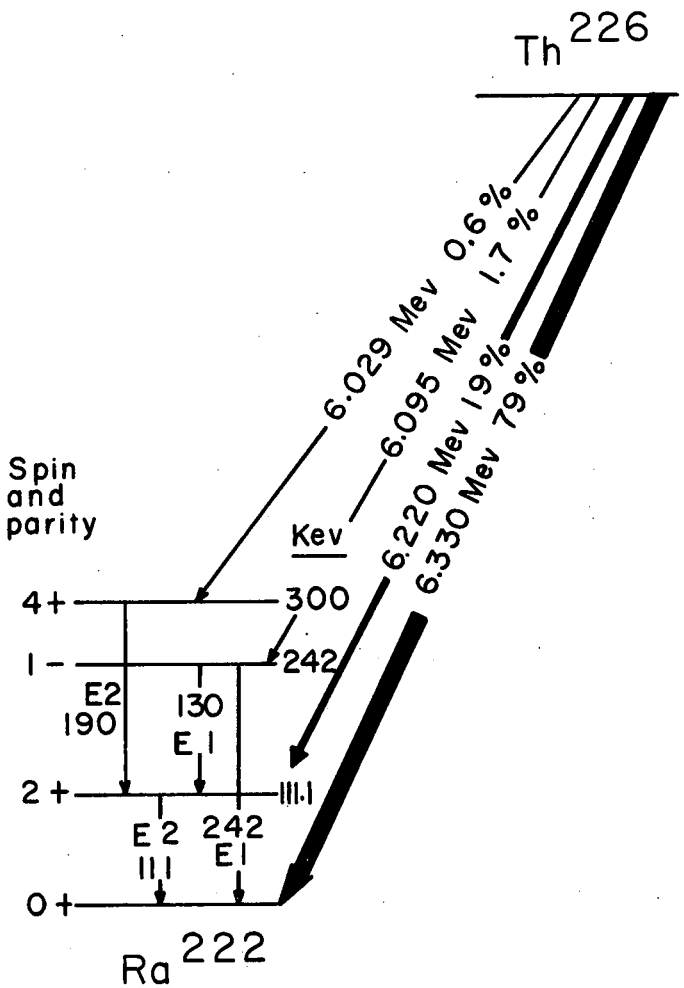

Fig. 8.2 Decay scheme of Th^{226}.

8.2.5 Thorium-227 (Radioactinium). Radioactinium, the direct decay product of actinium was found by HAHN[1] in 1906. HAHN was led to this discovery by noting that the regrowth of actinium X into an actinium sample freed from actinium X was much less rapid than was to be expected if actinium were transformed directly into this activity. This observation suggested the existence of an intermediate activity whose half-life Hahn found to be 19.5 days.

Growth curves showing the relationship of radioactinium to its parent, Ac^{227}, and to its descendants appear in Chapter 6. (See figures 6.40 and 6.41).

Radioactinium is an alpha emitter whose half life according to a recent redetermination is 18.17 days.[2] The alpha spectrum is exceedingly complex. Already in studies summarized by LEWIS AND BOWDEN[3] in 1934, eleven distinct alpha groups had been found. ROSENBLUM, VALADARES, PEREY AND GUILLOT[4] restudied the spectrum in 1952 and obtained a somewhat revised list of energies and intensities. The alpha spectroscopy of radioactinium is complicated by the close spacings of the groups and by the rapid ingrowth of the daughter, Ra^{223}, which also has a complex alpha spectrum in the same energy region. Later measurements were made by HUMMEL[5] and by PILGER[6]. We show the spectrum obtained by the latter in figure 8.3 (a) and list his results in Table 8.1. Pilger confirmed the main features of the spectrum reported by the ROSENBLUM group and found additional groups through higher resolution techniques. In all, he reported fifteen alpha groups. Such a complex spectrum is possible because a large number of excited levels are present in the daughter nucleus in the 0-400 kilovolt region and because alpha decay is for some reason highly hindered to the lowest-lying levels.

Since the alpha decay of Th^{227} leaves an appreciable number of daughter Ra^{223} atoms in 15 different excited states, it is quite understandable that the gamma ray and conversion electron spectrum of Th^{227} should be exceedingly complex. FRILLEY[7] reported nine gamma rays from studies made with a crystal spectrometer. In later studies of the conversion electrons FRILLEY, ROSENBLUM, VALADARES AND BOUISSIERES[8] fround evidence for 14 gamma rays between 30 and 335 kilovolts energy. PILGER recorded over 140 conversion electrons corresponding to more than 26 gamma rays lying in this same range of energies. The energies and intensities of these gamma rays are listed in the table.

PILGER[6] has constructed the decay scheme shown in figure 8.3 (b). Even with the precise knowledge of 15 excited levels of Ra^{223} given by the alpha groups all of the known gamma ray transitions cannot be placed with certainty in the decay scheme. From the viewpoint of the unified model of nuclear structure,

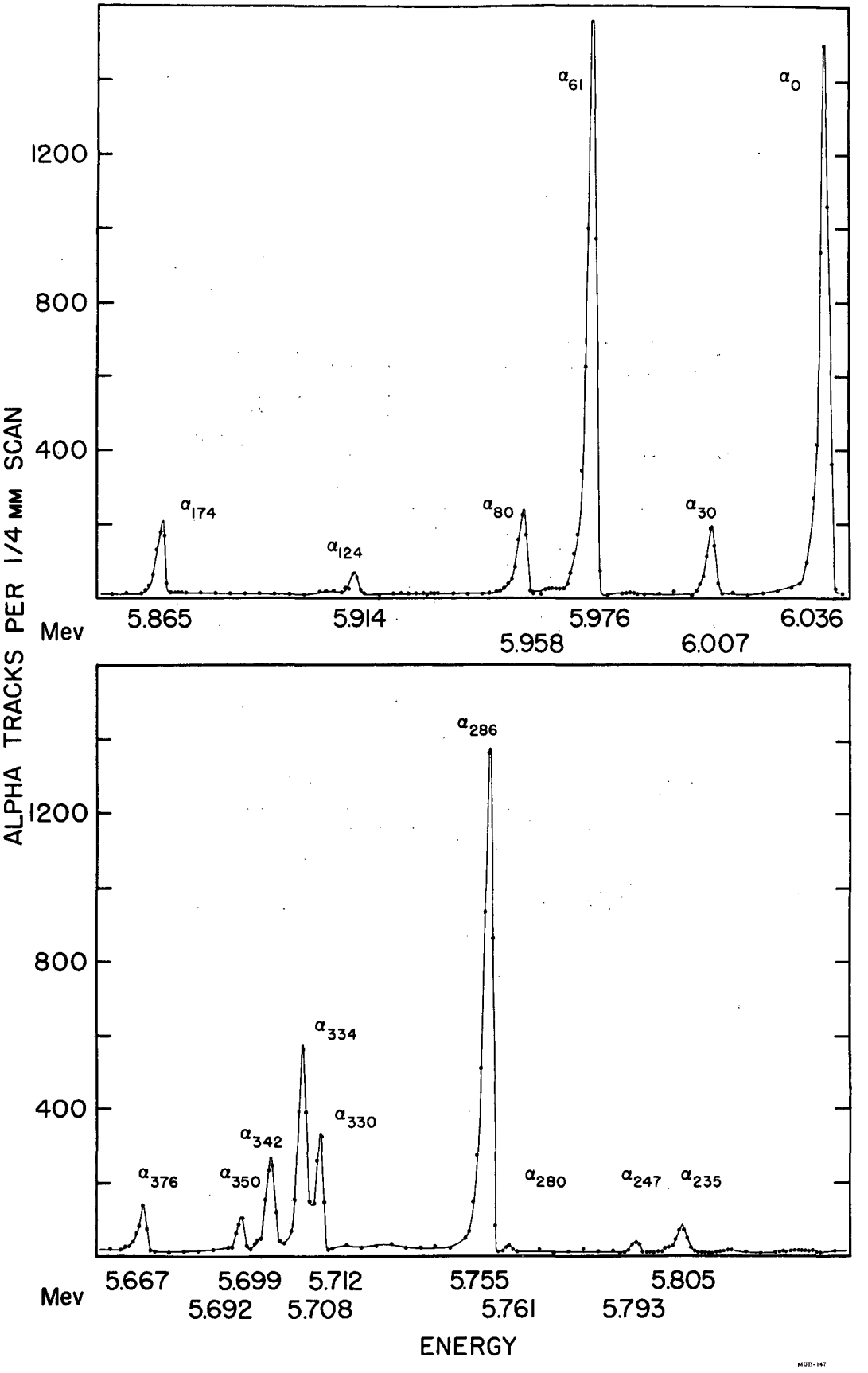

Fig. 8.3 (a) Alpha decay of radioactinium. The alpha spectrum as recorded photographically in a magnetic spectrograph of high resolution by Pilger.

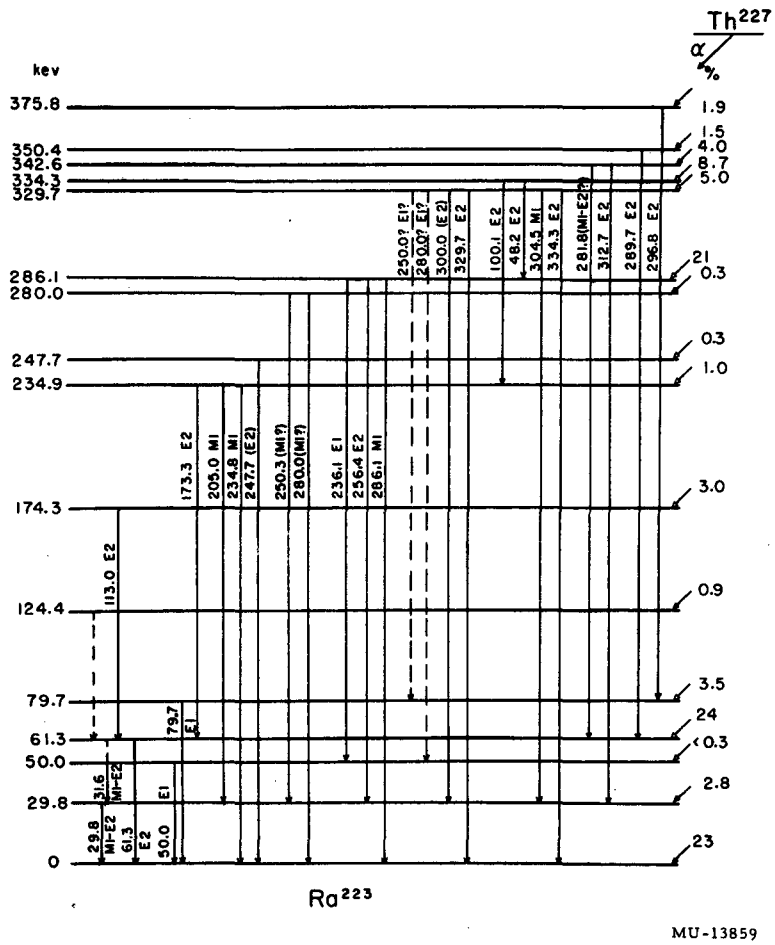

Fig. 8.3 (b) Alpha decay scheme of radioactinium as drawn by Pilger

Table 8.1

The Alpha Groups of Radioactinium (Th^{227})

Energy(Mev)	Decay Energy to ground	Percent Abundance
6.036*	0.0	23
6.007	30.0	2.8
5.976	61.5	24
5.958	79.9	3.5
5.914	124.2	0.9
5.865	174.4	3.0
5.805	234.9	1.0
5.793	247.1	0.3
5.761	280.0	0.3
5.755	286.1	21
5.712	329.8	5.0
5.708	334.2	8.7
5.699	342.6	4.0
5.692	350.4	1.5
5.667	376.0	1.9

*Based on 6.110 Mev energy for $Cm^{242}\alpha_o$

Results of R. Pilger

Q value 6.218

Internal Transitions in Decay of Th^{227}

Energy (kev)	Intensity (%)	Multipolarity
29.8	27	M1-E2
31.6	12	M1-E2
48.2	2.7	E2
50.0	13.6	E1(M2)
61.3	9.0	E2
79.7	4.6	E1
100.1	1.0	E2
113.0	4.2	E2
173.3	~1	E2
205.0	0.4	M1
234.9	2.0	M1
236.1	10.6	E1
247.7	weak	(E2?)
250.3		M1[a]
256.4	7.1	E2
280.0		M1[a]
281.8	~2	M1-E2?
286.1	1.5	M1
289.7	~0.9	E2?
296.8	~2.5	E2
300.0	~1	E2?
304.5	~2	M1(E2)?
312.7	3.5	E2
329.7	1.6	E2
334.3	5.2	E2

a. An E1 of the same energy probably also exists.

Data of Pilger[6]

the daughter isotope Ra^{223} lies in a transition group of nuclei between the strongly deformed transthorium nuclei, which show well-developed rotational bands, and the nuclei lying close to the double closed shell at Pb^{208}. It is difficult to classify nuclear levels in nuclei such as Ra^{223} which belong to this transition group. There is some evidence for rotational levels based on the ground state but the evidence is not conclusive.

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8.2.6 Thorium-228 (Radiothorium). Thorium-228 is a beta-stable alpha-emitter with a half-life[1] of 1.910 years. It occurs in nature as a member of the Th^{232} family. Its position in that family is discussed in Section 6.3 of Chapter 6. In the older literature of natural radioactivity thorium-228 bears the name radiothorium (symbol RdTh).

Isotopically pure samples of Th^{228} can be prepared from thorium ores or aged thorium compounds by isolating Ra^{228} (mesothorium[1]) and allowing Th^{228} to grow into the separated radium. For the success of this method the original radium-thorium separation must be exceptionally good. Thorium-228 may also be isolated from a U^{232} sample which has stood for some time; if the U^{232} is mixed with U^{233} there will be some contamination of the thorium daughter fraction with 7340 year Th^{229}. A third source of Th^{228} is the irradiation of radium or actinium with an intense flux of neutrons.[1]

$$Ra^{226}(n,\gamma)Ra^{227} \quad (\sigma = 18 \text{ barns}) \qquad Ra^{227} \xrightarrow[41 \text{ min}]{\beta^-} Ac^{227}$$

$$Ac^{227}(n,\gamma)Ac^{228} \quad (\sigma = 495 \text{ barns}) \qquad Ac^{228} \xrightarrow[6.13 \text{ hr}]{\beta^-} Th^{228}$$

The amount of contamination of the Th^{228} with the 18 day Th^{227} daughter of actinium depends on the neutron flux and the time elapsed between the pre- and post- bombardment separation of actinium and thorium.

The decay of Th^{228} to Ra^{224} is rather well studied. The alpha spectrum has been studied in some detail with a magnetic spectrograph[2-4] and several groups have measured the gamma-ray and conversion electron spectra.[4-8] The results of the most recent and thorough study are summarized in the decay scheme shown in the Figure 8.4.

Five alpha groups of Th^{228} have been identified, having energies and intensities as follows: 5.421 Mev (71%), 5.338 Mev (28%), 5.208 Mev (0.4%), 5.173 Mev (0.2%), and 5.137 Mev (0.03%). An upper limit of 0.01% has been set on the intensity of any lower energy alpha groups. The positions of these groups in the decay scheme are indicated in Fig. 8.4.

The gamma-ray spectrum of Th^{228} is well known and the results are summarized as follows: 84 kev (1.6%), 132 kev (0.16%), 167 kev (0.13%), 205 kev (0.03%), and 214 kev (0.30%). The intensity of any gamma ray of energy greater than 275 kev has been shown to be less than 0.001%. Coincidence measurements have established that the 132, 167, and 205 kev transitions

are all in coincidence with the 84 kev transition. The placement of the gamma rays in the decay scheme is very straightforward. The agreement between the alpha-particle and gamma-ray spectra is usually good in this case. In addition, the L conversion lines of the 84 kev transition have been studied and these indicate rather conclusively that this transition is E2. The total conversion coefficient of this gamma ray supports this assignment as does the lifetime of the 84 kev level, which has been measured as 7.6×10^{-10} seconds.

Radium-224, the daughter of Th^{228} alpha decay, lies on the edge of the region of nuclei in the heavy elements that have stable spheroidal deformations. This region extends from mass number approximately 220 up to the heaviest elements yet discovered. The fact that Ra^{224} lies just within this region means that a rotational band based on the ground state is still present, but that the equation $E_I = \frac{\hbar^2}{2\Im} I(I+1)$ is no longer exactly valid. The ground state rotational band in Ra^{224} is apparent in the Figure 8.4, the 2+ and 4+ members lying at energies 84.5 and 253 kev respectively. The 4+ assignment of the 253 kev level cannot be considered certain, but seems very likely because (1) the 167 kev gamma ray appears to be E2 from its total conversion coefficient, (2) there is no cross-over transition from the 253 kev level to ground, and (3) the systematics of other even-even nuclei in this region strongly suggest a 4+ level at approximately this energy. The value of $\frac{\hbar^2}{2\Im}$ in the equation above, as calculated from the energy of the 84.5 kev level, is 14.1 kev. This is considerably larger than is found for nuclei located well within the heavy-element spheroidal region, where $\frac{\hbar^2}{2\Im}$ is nearly constant and equal to ~ 7 kev. The larger value $\frac{\hbar^2}{2\Im}$ for Ra^{224} indicates a smaller moment of inertia, $\Im$, which presumably is the case because the spheroidal deformation is not so large as it becomes for heavier nuclei. If one uses the value of 14.1 kev for $\frac{\hbar^2}{2\Im}$ and calculates the energy of the I = 4 member of the band, this energy is 282 kev, almost 30 kev larger than is found. This deviation from the calculated value is characteristic for nuclei near the edge of regions of spheroidal deformation and in such regions higher terms must be added to the equation above. The second term $CI^2(I+1)^2$, where C is a constant, will necessarily give agreement with the data on Ra^{224}, since only two energy spacings are known. In cases where more levels are seen, it is sometimes also necessary to add a third term.

Of the two remaining levels in Ra^{224}, the one at 217 kev has been assigned spin and parity 1-, with a K value of 0. The spin and parity assignments are made on the basis of conversion coefficient arguments and angular correlation data. The earlier conversion coefficient arguments are simply that the sum of the intensities of the 214 and 132 kev gamma rays is 0.46%, and when this is compared with the alpha population of the 217 kev level, 0.4%, it is clear that the conversion coefficients of the gamma rays must be quite small. On this basis it was argued that the gamma rays are very likely E1, and since one of them terminates at the ground, 0+, level, the spin of the 217 kev level must be 1-. The angular correlation data[10] are more definite and show unambiguously that the spin and parity must be 1-. The assignment of the K value as 0 is made because the ratio of the intensities of the two gamma rays de-exciting the level is that predicted by the collective model of the nucleus if K = 0. The model predicts that the ratio of the reduced transition probability for the E1 gamma ray to the 0+ state divided by that to the 2+ state should be 2 if K = 1 and 0.5 if K = 0. The observed value is 0.44, rather clearly indicating a K value of 0. A number of such 1- (K = 0) levels have been found at low excitation energies in just this region of the periodic table. It so happens that the 217 kev level in Ra^{224} is the lowest energy at which such a state is known to occur. These states are generally ascribed to octupole vibtations of the nucleus, but why they occur at such low energies only in the region of radium is not yet fully understood.

The level at 289 kev in Ra^{224} has been suggested to have spin and parity 3- by the following reasoning. A comparison of the intensity of the 205 kev photons with the alpha population to the 289 kev state shows that the conversion coefficient of this transition must be very small and therefore the probable assignment is E1 (E2 is also possible). If the E1 assignment is correct, the spin of the 289 kev level must be 1-, 2-, or 3-. A spin of 2- is ruled out since the state receives direct alpha population, and this is not possible from a 0+ parent to a 2- final state. A spin of 1- seems unlikely since no cross-over transition to the ground state is observed. The spin of 3- is not unexpected, as the rotational band based on the 1- (K = 0) state has been found in other nuclei (and is expected) to have members 1-, 3-, 5-, ····. Thus the levels of Ra^{224} can be accounted for in terms of two rotational bands, one based on the ground state, and the other based on the

1- state, which, itself, is presumably due to an octupole vibration of the ground state configuration.

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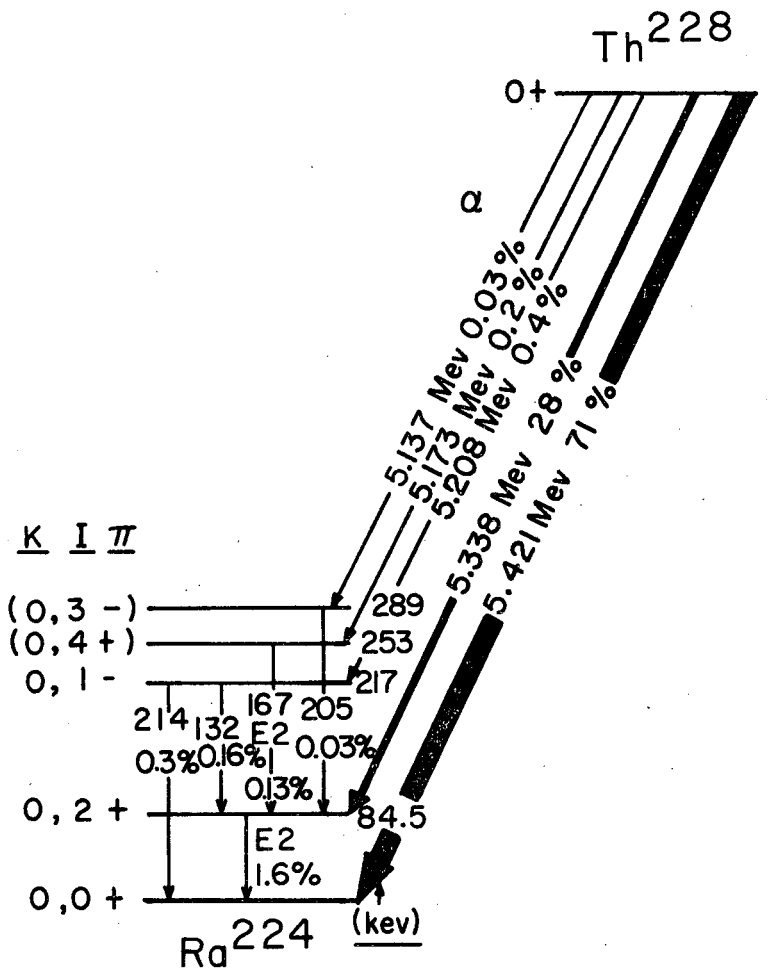

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Fig. 8.4 Decay scheme of Th^{228}.

8.2.7 Thorium-229. This isotope is a member of the neptunium ($4n + 1$) series[1,2] where it appears as the daughter product of U^{233}. The discovery experiments and the family relationships are discussed in Chapter 7 (See Section 7.1). Thorium-229 is an alpha emitter with a half-life of 7340 years.[2]

The alpha spectrum was measured in a magnetic spectrometer of high resolution by GOL'DIN, NOVIKOVA, PIROGOVA, AND TRETIAKOV[3] and in a gridded ion chamber and in a solid state detector, both of moderate resolution, by ENGELKEMEIR AND GINDLER.[4] The results of these two studies are compared in Table 8.2. The agreement is good for the more intense groups. There appears to be a systematic difference in the calibration of the energy scale by the two laboratories since the reported values for the highest energy alpha groups differ by 3 kev and for the lowest energy groups by 17 kev.

Both research groups report preliminary studies of the complex gamma spectrum. GOL'DIN et al.[3] report well established transitions with energies 17.2, 42.8, 69.9, 75.5, 137.2, 156.6, 193.4, and 210.5 kev. Others are reported with less certainty at 29.1, 31.6, 56.8, 58.9, 85.0, 132.1, 154.4, 179.6, and 242.0 kev. ENGELKEMEIR AND GINDLER[4] report L and K x-rays and gamma-rays with energies 25.3, 27.9, 31.5, 44.3, 75.1, 86.2, 136.7, 152.4, 193.7, and 210.2. Neither group in their original study had been able to construct a detailed decay scheme, but a few features were mentioned.

The favored alpha decay proceeds to the Ra^{225} daughter level at 214 kev. Since the ground state of Th^{229} has spin 5/2 and the Nilsson wave function assignment 5/2+ [633], for reasons which are discussed in the U^{233} report (See Section 8.4.6), the Nilsson assignment of the 214 kev level in Ra^{225} may also be 5/2+ [633]. There is some evidence for rotational structure based on this level but the level spacing does not follow the I(I+1) rule. There is a compression of the rotational spacings caused by the interaction with near-lying Nilsson states of similar K value. The close spacing of Nilsson levels and the distortion of rotational spacings is observed for many nuclei in this range of mass numbers.

The alpha transition of the highest observed energy is greatly hindered and there is evidence that the true ground state transition may be even higher in energy and in hindrance. ENGELKEMEIR AND GINDLER[4] report that L_α, L_β, and L_γ x-rays are in coincidence with the 5.051 Mev alpha group. This result places the ground state of Ra^{225} at least 19 kev below the level populated by the 5.051 Mev alpha particles.

Table 8.2

Alpha Groups of Th^{229}

ENGELKEMEIR AND GINDLER (1960)				GOL'DIN AND CO-WORKERS (1959)			
E_α (kev)	Energy Ra^{225} state (kev)	Intensity (%)	α Hindrance	E_α (kev)	Energy Ra^{225} state (kev)	Intensity (%)	α Hindrance
>5070	---	<0.2	>9000	---	---	---	---
5051	0	8.0	220	5048	0	6.7	330
---	---	---	---	5028	20	≃0.2	$\sim 10^4$
---	---	---	---	5003	45	≃0.1	$\sim 10^4$
4975	77	5.6	110	4.971	78	3.4	200
4966	87	5.7	90	4.961	88	6.0	100
---	---	---	---	4.925	125	0.25	~1000
4901	153	10.6	18	4.894	156	10.7	25
4845	210	56.1	1.5	4.837	214	58.2	1.5
4814	241	10.1	5.2	4.806	246	11.4	7
4797	259	2,0	20	4.788	264	1.0	40
4763	293	1.5	16	4.751	302	1.5	20
4695	363	0.4	20	4.678	376	0.4	25
4608	451	0.05	40	---	---	---	---
4480	581	0.03	8	---	---	---	---

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2. F. Hagemann et al. Phys. Rev. 72, 254 (1947), a preliminary note; and Phys. Rev. 79, 435 (1950).

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5. G. E. Kocharov, A. P. Komar, G. A. Korolev, I. N. Marov, and Yu. A. Surkov, Izvest. Akad Nauk S. S. S. R. Ser. Fiz. 23, 855-8 (1959) July.

8.2.8 Thorium^{230} (Ionium). Thorium-230 is an alpha-emitter with a half-life of 8.0×10^4 years.[1] It is a member of the naturally occurring U^{238} family, (See Section 6.2 of Chapter 6), and may be prepared simply by separation from uranium ores. For samples of high isotopic purity, however, the Th^{230} must be mass separated from the Th^{232} impurities present in the ores. Owing to the low specific activity of Th^{230}, high-resolution alpha-particle and conversion-electron spectroscopy have been difficult. Nevertheless the decay scheme has received rather extensive study, and is, for the most part, well established.

The alpha-particle spectrum of Th^{230} has been studied by several groups[2-5] using both magnetic spectrographs and ionization chambers. There is not complete agreement among the experiments at the present time; however, at least four alpha groups seem to be clearly established. These have energies and intensities as follows: 4.682 Mev (76%), 4.615 Mev (24%), 4.476 Mev (0.12%), and 4.437 Mev (0.03%). (See Table 8.3). The placement of these groups in the decay scheme is indicated in Fig. 8.5 by the solid lines. The dashed lines indicate alpha groups inferred from the gamma ray studies.

Eight gamma rays accompanying Th^{230} decay have been found from studies of the singles gamma-ray spectrum and the gamma-gamma coincidences.[6-9] The gamma rays as given in one of the most recent studies[7] are: 68 kev (0.59%), 110 kev (1×10^{-4}%), 142 kev (0.07%), 184 kev (1.4×10^{-2}%), 206 kev ($\sim 5 \times 10^{-6}$%), 235 kev ($\sim 5 \times 10^{-6}$%), and two gamma rays of 253 kev having intensities of 1.7×10^{-2}% and $\sim 8 \times 10^{-4}$%. An upper limit of 7×10^{-6}% has been set on any radiations between 300 and 700 kev. Of the observed gamma rays, only four can be seen in the singles spectrum. These have energies of 68, 142, 184, and 253 kev; and it was further shown that of these the 142 and 184 kev transitions are in coincidence with the one of 68 kev. These four gamma rays can easily be fit into the decay scheme established by the alpha spectrum as shown in the Figure 8.5. Next it was shown that a part of the 253 kev photons were in coincidence with a 68 kev transition, indicating a new level at 320 kev. An experiment aimed at deciding whether (1) another gamma ray of 253 kev was present in coincidence with the established 68 kev transition, or (2) another 68 kev gamma ray was present in coincidence with the established 253 kev transition, indicated that the former, (1), is probably the correct situation. Thus the second 253 kev transition is characterized. A 110 kev transition in

coincidence with the 142 kev gamma ray was also found to de-excite the new 320 kev level. Finally, also in coincidence with the 142 kev gamma ray two very low intensity transitions of energy 206 and 235 kev were found. These were interpreted as establishing the levels at 416 and 445 kev. The reasoning behind these placements of the two weak transitions is discussed in more detail below.

Only the 68 kev transition has been seen in the conversion electron spectrum of Th^{230}.[6] The energy of this transition has been placed as 67.7 kev from these studies and, also, since the conversion was found to be principally in the L_{II} and L_{III} subshells, an E2 assignment can be made. This assignment is confirmed by the total conversion coefficient of the transition (found by comparing the 68 kev photon intensity with the alpha population to the 68 kev level)[8] and by the lifetime of the 68 kev level, which has been measured to be 6.3×10^{-10} sec.[10] Thus the 68 kev level must have spin and parity assignments 2+, as is found generally for the first excited state of an even-even nucleus.

As is indicated in Fig. 8.5, the levels of Ra^{226} have been interpreted in terms of two rotational bands based on the ground, 0+, state and the 1- state. In the ground state band, spins of 2+ and 4+ have been clearly established for the 67.7 and 210 kev levels. This has been done by a combination of conversion coefficient and angular distribution arguments.[8,11-13] Both gamma-gamma and alpha-gamma angular distribution measurements have been made to establish the spin of the 210 kev level, and this assignment is one of the few really proven 4+ assignments for the third member of the ground state rotational band. In many other cases the spin assignment of this level rests largely on the agreement with the Bohr-Mottelson formula:

$$E_I = \frac{\hbar^2}{2\Im} I(I+1) \qquad (1)$$

The assignment of the 6+ member of the ground state band is by no means so clearly established. Here the arguments are (1) the decay only to the 4+ state is consistent with a spin of 6+, and (2) the systematic trends in energy levels of other even-even nuclei in this region suggests that there should be a 6+ level in Ra^{226} at about this energy. These arguments are not conclusive and the parentheses around the spin in Fig. 8.5 indicate that the assignment is tentative.

Because Ra^{226} lies rather near the edge of the region of spheroidal nuclei in the heavy elements (this region extends from about mass 220 up to

the heaviest elements yet discovered) significant deviations occur from equation (1). In order to fit the three spacings experimentally observed, a second term, $BI^2(I+1)^2$, and a third term $CI^3(I+1)^3$ must be added. In this case the constants $\frac{\hbar^2}{2\mathfrak{I}}$, B, and C may be evaluated to be: 11.74 ± 0.10 kev, 0.080 ± 0.010 kev, and 0.00085 ± 0.00025 kev respectively. Since there are only three spacings, it is necessary that three constants will fit the experimental data, and the fit obtained indicates nothing about the physical validity of such an expansion in terms of powers of $I(I+1)$. Also the limits of error on the constants are those resulting from uncertainties in the energies of the levels, and it is quite possible that, when more spacings are known, a $DI^4(I+1)^4$ term will be found necessary, and the addition of this term will no doubt affect the value of C, perhaps so much that it will lie outside the indicated limits of error.

The level in Ra^{226} at 253 kev has been shown to have spin and parity 1-, and a K value of zero. The 1- assignment is based on angular distribution measurements (the 0+ $\xrightarrow{\alpha}$ 1- $\xrightarrow{\gamma}$ 0+ pattern is very distinctive) and is quite conclusive.[14] The K = 0 assignment is made because the reduced transition probability of the transition from this level to the 0+ state divided by that to the 2+ state is 0.47; whereas the unified model predicts a value of 2 for this ratio if K = 1 and 0.5 if K = 0. A number of such low-lying 1-, (K = 0) states have been found in this region of the periodic table, and they are generally ascribed to octupole vibrations of the ground state configuration.

The 3- and 5- spin assignments (both with K = 0) are tentative and are based on the facts that (1) the decay of the levels is consistent with these assignments, (2) even spin states with odd parity could not receive direct alpha decay from a 0+ parent, and (3) such levels are expected to be present as rotational band members based on the 1- (K = 0) state. The K = 0 assignment for the 3- level is supported by the reduced transition probability (assuming the transitions to be E1) of the gamma ray to the 2+ level divided by that to the 4+ level, which is experimentally 0.7, and theoretically 0.75. The low limit set on the abundance of the possible (320 kev) crossover transition from this level to the ground state pretty effectively rules out any spin lower than 3 for this level. Because of the small energy difference between the assigned 5- and 6+, one expects to see only the 5- ——> 4+ transition if K = 0, as is observed.

If the 320 and 445 kev levels do, indeed, comprise the rotational band based on the 1- state, this band has two interesting features. First, the value of $\frac{\hbar^2}{2\mathfrak{I}}$ is 6.7 kev for this band compared with 11.7 kev for the ground state band, and second, there is no deviation observed from equation (1), compared with large deviations for the ground state band. Both of these features can possibly be explained by a rather large mixing of this I = 1, K = 0 band with the other expected octupole vibrational bands, specifically in this case with the I = 1, K = 1 band. It will be interesting to see if such K $\neq$ 0 octupole bands occur systematically in the even-even nuclei of this region.

The 1- level of Ra^{226} plays a prominent role in the electron capture decay of Ac^{226}. This is illustrated in Figure 8.6. A study of the decay of Ac^{226} has helped to confirm the assignment of the level at 253 kev.[15]

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Table 8.3

Alpha Groups of Th^{230} (Ionium)

Alpha particle energy* (Mev)	Alpha intensity (%)	Alpha decay hindrance factor	Final state	
			Energy (kev)	spin and parity
4.682	76	(1)	0	0+
4.615	24	1.1	67.76	2+
4.476	0.12	12	210	4+
4.437	0.03	38	253	1-
(4.368)	0.001	370	320	3-
(4.273)	$\sim 5 \times 10^{-6}$	8200	416	6+
(4.245)	$\sim 5 \times 10^{-6}$	4900	445	5-

*Alpha groups in parenthesis were detected only indirectly via γ-ray measurements.

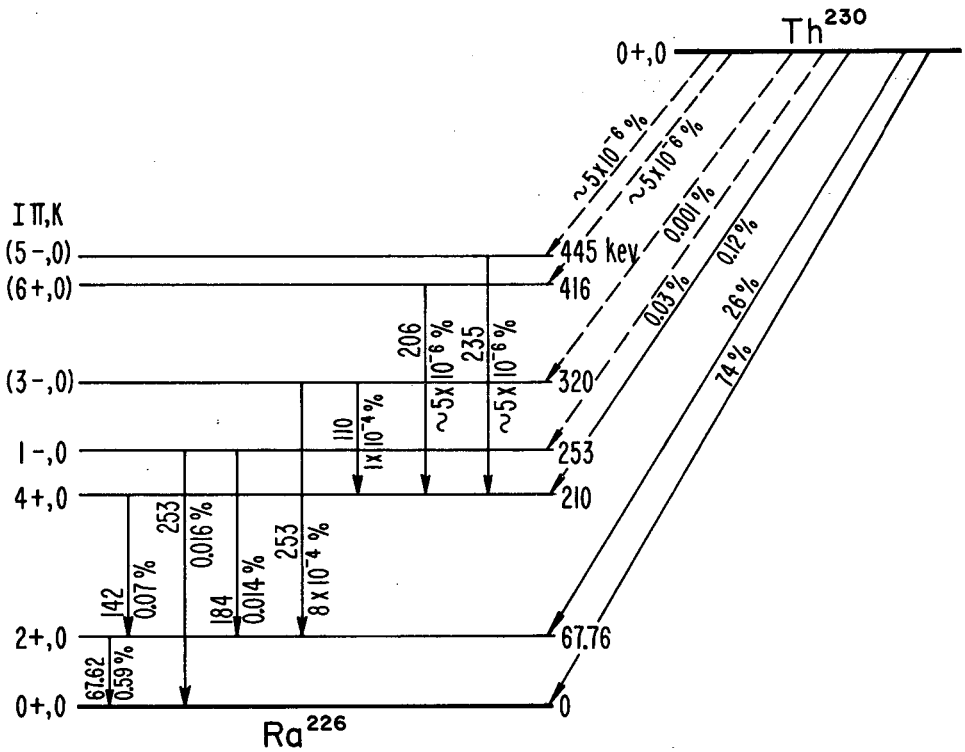

Fig. 8.5 Decay scheme of Th^{230} (Ionium) as drawn by Stephens, Asaro, and Perlman. Note that the observed levels of Pa^{226} are interpreted as a rotational band with 0+, 2+, 4+, and 6+ spin states based on an I = 0 K = 0 ground state and a second rotational band with 1-, 3-, and 5- spin states based on an I = 1 K = 0 fundamental state.

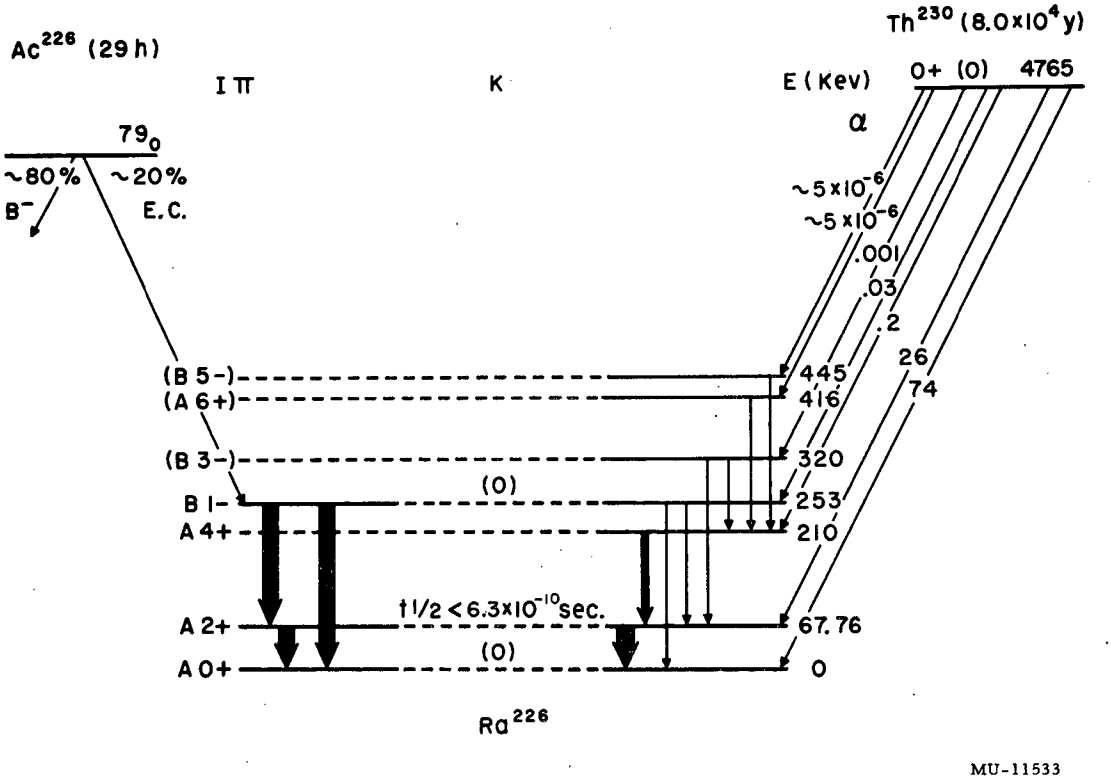

Fig. 8.6 Levels of Pa^{226} observed in the electron capture decay of Ac^{226} and the alpha decay of Th^{230} (ionium). The vertical arrows representing the experimentally observed gamma transitions indicate qualitatively by their width the relative transition intensities. In the Ac^{226} decay scheme the Pa^{226} levels are dotted except for those which participate in the decay of Ac^{226}.

8.2.9 Thorium-231 and Uranium-231. Thorium-231, a beta-emitter of 25.6 hours half-life is well known as the "UY" of classical radioactivity, the daughter product of U^{235} decay. Its early history and its relationship to the U^{235} decay chain are discussed in Chapter 6. Thorium 231 can be isolated from U^{235} sources (preferably enriched U^{235} sources) or it can be conveniently made by the neutron irradiation of ionium.

$$Th^{230}\ (n,\gamma)\ Th^{231}$$

The beta decay of Th^{231} has been studied by several groups. There are a number of older papers in the literature reporting the radiations of Th^{231} as determined by absorption curve methods or by nuclear spectrometers of low resolution. References to this literature are made in a paper by JAFFEY, LERNER, AND WARSHAW.[1] In recent years nuclear spectrometers of high resolution have been used to investigate the moderately complex decay of Th^{231}.[2-6] Particularly important has been the measurement of conversion electrons in permanent magnet spectrometers of 0.1 percent resolution. The analysis of the radiations and the decay scheme given here are based chiefly on a paper by HOLLANDER, STEPHENS, ASARO, AND PERLMAN.[6] It is convenient to discuss the electron capture decay of U^{231} at the same time since both U^{231} and Th^{231} decay to the same daughter nucleus and many of the same Pa^{231} gamma transitions are seen in the decay of both nuclides. Uranium-231 is a 4.2 day activity prepared by bombardment of Pa^{231} with deuterons, protons or helium ions.

The photons emitted by Th^{231} and U^{231} were studied[6] with a NaI (tℓ) scintillation detector coupled to a 50-channel pulse-height analyzer with the results shown in Table 8.4. This analysis was useful for obtaining the photon intensity of several of the important gamma rays but could not reveal the true complexity of the gamma spectrum. The permanent magnet spectrographs were able to reveal the conversion electrons of the numerous gamma rays which are also listed in Table 8.4. In the case of U^{231} a prominent spectrum of Auger electrons was seen.[6] The electron energies are listed in Table 8.5. The multipolarity assignments were made chiefly on the basis of the comparisons of the L- or M-subshell conversion electron ratios with theoretical predictions for the various multipoles. In some instances the absolute values for the conversion coefficients were used to select or to corroborate the multipolarity.

Table 8.4

Gamma Transitions in Decay of Th^{231} and U^{231}

Decay of Th^{231}					Decay of U^{231}		
Measured Photon Energy (kev)	Photon Intensity per 100 Disintegrations	Transition Energy from Conversion Electrons (kev)	Multipole Order	Total Transition Intensity	Measured Photon Energy (kev)	Photon Intensity per 100 Disintegrations	Transition Energy from Conversion Electrons (kev)
17 (L x-rays)		17.21			17 (L x-rays)		
		18.07					18.05
26±2	12.5±2	25.65	E1	73%	26	12	25.64
		58.53	E2	74%			58.54
		63.8					
		68.5					68.5
		76.1					
		81.16	M1	10%			81.3
	4±2	82.01	M1	8%			82.1
84±3	7.2±1	84.17	E1	23%	84	7	84.18
		89.8					
95±4 (includes x-rays)	2.4±0.5	99.28			93 (includes x-rays)		103.2
		135.8		~0.3%			
140	0.2						
		146.1		~0.3%			
160	0.2	163.3	M1	1.5%			
~180	~0.06						
218±3	0.05				220±4	~1	
~310	0.004						

Data taken from Hollander, Stephens, Asaro, and Perlman, Ref. 6. In the case of Th^{231} data on many of these gamma rays was reported by Mize and Starner[2] and by Freedman, Jaffey, Wagner, and May.[3]

Table 8.5

Auger Electrons from U^{231} Decay

KLX Transitions	ENERGY (exp) Kev
$K\ L_I L_I$	70.05
$K\ L_I\ L_{II}$	70.87
$K\ L_I\ L_{III}$	74.45
$K\ L_{II} L_{II}$	---
$K\ L_{II}\ L_{III}$	75.21
$K\ L_{III}\ L_{III}$	78.78
$K\ L_I\ M_I$	85.88
$K\ L_I\ M_{II}$	86.29

From Hollander, Stephens, Asaro, and Perlman.[6]

Two sodium iodide crystal detectors were used in a coincidence arrangement to detect gamma ray photons emitted within a period of 5 microseconds of each other.[6] One important fact brought out by this work was that the 26 kev and the 84 kev transitions are not in coincidence with each other, and that the high energy spectra coincident with both are identical. These facts suggest that the 26 kev and the 84 kev transitions originate at the same Pa^{231} level. This conclusion is strengthened by the fact, discovered by coincidence experiments performed with apparatus of greater time-resolution ability, that the 26 kev and 84 kev have measurable and identical half lives. STROMINGER AND RASMUSSEN[4] report a value of 4.1×10^{-8} seconds for this half life while MIZE AND STARNER[2] report 4.5×10^{-8} seconds. HOLLANDER, STEPHENS, ASARO, AND PERLMAN[6] measured the half life of the 84 kev transition in the decay of U^{231} and found a value of 4.1×10^{-8} seconds.

Gamma-gamma coincidence measurements were made on U^{231} with K x-rays as gate pulses.[6] These revealed the 26- and 84-kev photons in abundances of about 12 and 7 percent, respectively, per K x-ray gate pulse. The 220 kev photon was found to be in coincidence with both 26 and 84 kev radiation.

Beta-gamma measurements were made in the case of Th^{231} decay with a double detector system consisting of an anthracene crystal and a NaI (tℓ) crystal. These were useful chiefly for setting the abundance of the 25.6 kev photon as 12.5 ± 2% of the beta decay events and, by indirect estimates, for setting the abundances of several other gamma rays. These abundance values are given in Table 8.4. Another important fact established by the beta-gamma experiments was that the highest energy beta group was in coincidence with 84 kev photons.

The beta spectrum of Th^{231} was studied by FREEDMAN, JAFFEY, WAGNER, AND MAY[3] who observed three beta groups: 302 kev (44%), 216 kev (11%) and 94 kev (45%). JULIANO[5] obtained a somewhat different resolution as follows: 299 kev (39%), 218 kev (33%), 134 kev (20%) and 90 kev (8%).

HOLLANDER AND CO-WORKERS[6] summarized their measurements on Th^{231} and U^{231} in the decay scheme reproduced here as Figure 8.7. This scheme differs substantially from those suggested earlier by other authors. We mention here some of the arguments regarding the placement of levels, but refer the reader to the original paper for detailed justification.

The following facts establish clearly that there are levels in Pa^{231} at 58.5 and 84.2 kev.

1. The transition sum 58.53 + 25.65 equals 84.18 is in excellent agreement with the measured crossover energy 84.17 kev.
2. The 26 and 84 kev photons have the same half life.
3. The 58 kev has been excited in Pa^{231} target samples by the Coulombic excitation process.[7]

Three other sets of gamma energy sums were regarded as significant in the construction of the decay scheme.

$$18.07 + 81.16 = 99.23$$
$$17.21 + 82.01 = 99.22$$
$$\text{crossover} = 99.28$$
$$146.1 + 17.2 = 163.3$$
$$\text{crossover } 163.3$$

The ground state spin of Pa^{231} is 3/2 as measured by hyperfine structure analysis.[8]

We turn now to a discussion of the assignment of Pa^{231} levels to Nilsson orbitals. It is convenient in this connection to examine the Nilsson diagram in which the calculated energies of single particle orbits in a spheroidal potential are plotted as a function of a nuclear eccentricity. In the case of Pa^{231} the important Nilsson states are those available to the 91-st proton. A diagram showing the Nilsson states for proton number greater than 82 is located in Chapter 3.

The state labeled 1/2 - [530] is suggested by the Nilsson diagram, and HOLLANDER, STEPHENS, ASARO, AND PERLMAN[6] select it for the ground state of Pa^{231}. When the K-quantum number is 1/2 the energies of rotational states are given by the formula

$$E_i = \frac{\hbar^2}{2\,\Im}\,[I\,(I+1) + a\,(-1)^{I+1/2}\,(I + 1/2)]$$

in which

$\hbar$ is Planck's constant divided by 2π
$\Im$ is the moment of inertia
I is the spin of the state, and
"a" is a decoupling parameter which takes account of the partial decoupling of the k = 1/2 odd particle from the collective motion.

This formula is discussed in Chapter 3. If "a" happens to be < -1 an interesting inversion of the order of rotational states can occur; the 3/2

level can drop below the 1/2, the 7/2 below the 5/2, etc. Apparently, just this situation occurs in Pa^{231} so that the ground state (with measured spin 3/2) is the 3/2 member of a K = 1/2 band. Similarly, the 7/2- level lies below the 5/2 level.

The same 1/2- [530] Nilsson assignment has been made to the ground state of Pa^{233} from an analysis of decay scheme data for Np^{237} (See Section 9.1.9 of Chapter 9). The same inversion occurs in the ground state rotational band of Pa^{233} so that the lowest-lying level is the state with spin 3/2. In this case also the 7/2 level lies below the 5/2 level of rotational excitation. Since Pa^{231} and Pa^{233} both have 91 protons this similarity in the lowest-lying levels is natural.

The 84.1 kev level in Pa^{231} decays by electric dipole transitions to the $3/2^-$ ground state and the 7/2- state at 58.5 kev. Thus the 84.1 kev level has even parity and spin 5/2. An assignment of Nilsson quantum numbers to the 84 kev level is easily made since the 5/2+ state labeled [642] lies immediately above state [530] on the Nilsson diagram. It is consistent with this interpretation that the ground states of Np^{237} and Np^{239} with two additional protons have also the configuration 5/2+ [642].

There is no direct beta decay observed from Th^{231} to the ground state rotational band of Pa^{231} in spite of the fact that spin states 3/2, 1/2, 7/2, and 5/2 are available. This fact is attributed to the operation of the K-selection rule which states that ΔK must not exceed the multipolarity, ΔL, of the beta transition. The ground state Nilsson orbital assignment for Th^{231} (141 neutrons) is 5/2 + [633]. This assignment is also made to the ground state of U^{233} which also has 141 neutrons. The beta spectrum analysis given by FREEDMAN AND CO-WORKERS[3] or by JULIANO[5] cannot be reconciled in detail with the level system or transition abundances derived from the cited measurements on the gamma transitions, so the beta transitions are shown in incomplete form in Figure 8.7.

Uranium-231 decays chiefly to the 84 kev level and/or the 102 kev level of Pa^{231} although there is a very small direct population of the upper states. Uranium-231 has 139 neutrons. The state predicted from the Nilsson diagram for the 139th neutron is 5/2- [752]; this assignment is consistent with the observed ft value of 6.1, since the transition would be of the type forbidden ($\Delta I = 0$) unhindered.

The 84 kev transition which occurs in the decay of both Th^{231} and U^{231} is worthy of special comment. It was mentioned above that the half life of this transition has been found to be 4.1×10^{-8} seconds. This half life is very long compared to the predictions of "single-particle" transition probability formulas. In fact the photon retardation factor is 2.8×10^{6}. This E1 transition belongs to a group of E1 transitions with anomalously long half lives which occur in the odd mass isotopes of heavy elements. In this particular case the chief part of the retardation may be attributed to violation of the selection rule in K. In other cases the retardation has been qualitatively explained in terms of violations of selection rules in the asymptotic quantum numbers N, n_z, and Λ as is discussed in Section 3.5.7 of Chapter 3. These transitions also show anomalies in the L-shell conversion coefficients. In the case of the 84 kev E1 transition in Pa^{231} the L_{III} conversion coefficient agrees with theoretical calculations whereas the L_I and L_{II} are 21 and 15 times larger than the theoretical values. This interesting case and other similar anomalies in other E1 transitions are discussed critically in a paper by ASARO, STEPHENS, HOLLANDER, AND PERLMAN.[9]

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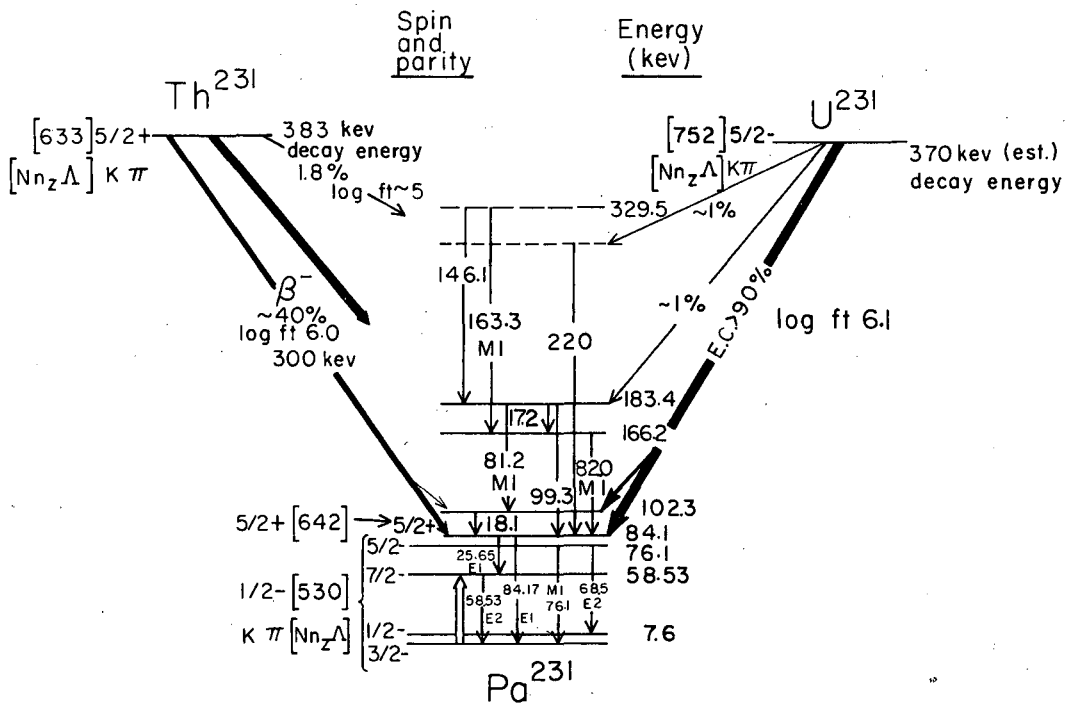

8.7 Decay schemes of Th^{231} and U^{231} as drawn by Hollander, Stephens, Asaro, and Perlman. The half-lives are 25.6 hours and 4.2 days, respectively.

8.2.10 Thorium-232. Natural thorium consists of the isotope, Th^{232}. This isotope gives rise to the thorium series of radioactive isotopes which is thoroughly discussed in Chapter 6. Determinations of the half-life of thorium are quoted in Table 6.11 of that chapter. The "best value" is 1.39×10^{10} years, which corresponds to a specific activity of only 246 alpha disintegrations per minute for each milligram. The main alpha-particle group has an energy of 4.007 Mev according to the ion chamber measurements of HARVEY, JACKSON, EASTWOOD, AND HANNA.[1] The intensity of the main group is about 76 percent. A second group appears at 55 - 65 kilovolts lower energy in about 24 percent abundance. KOCHAROV, KOMAR, AND KOROLEV[2] have also observed this alpha particle group in some careful ion-chamber measurements. This result is amply confirmed by the observation[3-5] of electron tracks paired with alpha particle tracks in nuclear emulsions impregnated with thorium salts. The electron tracks have the proper energy to be identified with the conversion of a 55 kev gamma ray. KOCHAROV, KOMAR, AND KOROLEV[2] also saw alpha particles in 0.2 ± 0.08 percent intensity which populate a level at 185 ± 5 kev in the Ra^{228} daughter nucleus. The 60 kev level and the 185 kev levels in Ra^{228} can be assigned with some confidence to the 2+ and 4+ states of rotational excitation of a non-spherical ground state. Some alpha decay may occur in even smaller intensity to other levels of collective excitation of the Ra^{228} daughter but the low specific activity of thorium makes it extremely difficult to look for these low-intensity transitions.

Excited levels in Th^{232} have been produced by the Coulombic excitation process. These levels are discussed in connection with the comments on U^{236} in Section 8.4.10 below. They are also mentioned in the discussion of the Unified model of the nucleus in Chapter 3.

Thorium-232 has an extremely long half life for spontaneous fission. PODGURSKAYA AND CO-WORKERS[6] set an upper limit of 10^{20} years. FLEROV AND CO-WORKERS[7] later raised this limit to 10^{21} years.

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8.2.11 Thorium-233. The short-lived Th^{233} is prepared by the neutron irradiation of Th^{232}. The discovery of Th^{233} is briefly described in Section 7.1 of the last chapter where the 4n + 1 series of isotopes is described. The half-life is usually quoted as 23.5 minutes, but a determination by JENKINS[1] led to the value, 22.12 minutes. Thorium-233 decays by emission of 1.24 Mev[2-5] beta-particles directly to the ground state of Pa^{233} in the majority of its disintegrations.

The work of FREEDMAN AND CO-WORKERS[5] on intense sources of Th^{233} prepared in a high-flux reactor has shown that the beta spectrum is complex and that many low intensity gamma rays accompany the decay. These authors find a main beta group at 1.245 ± 0.003 Mev in somewhat less than 87 percent abundance while the remaining beta transitions are divided among the following groups: 1.158, 1.073, 0.88, 0.79, and 0.58 Mev. In experiments using magnetic and scintillation spectrometers the gamma rays shown in Table 8.6 were found but these gamma rays have not been placed in a decay scheme. There should be some similarity in the gamma radiations of Th^{233} and Np^{237} since both these nuclides populate excited levels of Pa^{233}. The decay scheme of Np^{237} is discussed in Section 9.1.10 of Chapter 9. It can be noted that the 29.2 kev, 56.7 kev, and 86.9 kev radiations reported by FREEDMAN AND CO-WORKERS[4] in the decay of Th^{233} may correspond to the de-excitation of the first two excited levels of Pa^{233} which lie at 57 and 86 kev.

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Table 8.6

Gamma Radiations of Th^{233}

γ-ray Energy	γ's per Transition	Conversion electrons per transition
29.2	2.1%	5.9%
56.7	0	8.4%
86.9	2.7%	2.3%
171	0.7%	
195	0.3%	
253		
359		
453	1%	
590		
670	0.25%	
751		
895	0.14%	
+ higher		

Unpublished results of Freedman, Engelkemeir, Portor, Wagner, and Day.[5]

8.2.12 Thorium-234 and the UX_1 - UX_2 - UZ Complex. Thorium-234 is the daughter product of U^{238} and is readily isolated from uranium compounds by chemical separation of a thorium fraction. It bears the classical name of UX_1. The important role it played in the history of radioactivity and its genetic relationship to the rest of the uranium family of natural radioactivities are discussed in Chapter 6. We are concerned here solely with the details of the radiations of UX_1. Because the radiations of UX_2 and UZ are almost always associated with those of UX_1 it is convenient to discuss all three activities in one place. The relationships of the members of the UX complex to each other are shown in Figure 8.8.

Thorium-234 is a beta emitter with a half life of 24.10 days.[11] It has a two component beta spectrum consisting of a 191 kev group in 65 percent abundance and a 103 kev group in 35 percent abundance. Various investigations are summarized in Table 8.7. The gamma spectrum is also reported to be simple. Several investigators[3,4,8] have reported gamma rays of 91.4, 62.8, and 29.2 kev.

Beta-electron coincidence measurements by ONG[4] and HEERSHAP,[5] and gamma-beta coincidence measurements by DE HAAN[6] and JOHANSSON[8] show that the 91.4 kev gamma ray is in coincidence with the 103 kev beta particles. Since the other two gamma rays sum to $\sim$ 92 kev, they were thought at one time to be in cascade, with the 91.5 kev transition as the crossover. The multipolarity assignments on the basis of L shell conversion coefficients are M1 for the 91.5 kev transition and E1 for the 62.8 kev transition.[4] To conserve spin and parity the 29 kev transition must be E1. However WOOD[22] found too few photons of the 29 kev transition which means either that the L conversion coefficient is quite anomalous or that the transition is E2. If it is E2 then the 91.4 kev cannot be a crossover transition summing a 62.8 and 29.0 kev cascade. This suggests that other transitions, perhaps with nearly identical energies, are present in the decay of Th^{234}. STEPHENS[12] has found some preliminary evidence that this indeed is the case.

The isomeric species UX_2 (1.17 minutes) and UZ (6.7 hours) were discovered in 1913 and 1921 and numerous investigations of them have been made in past decades particularly in the last two. Nonetheless there has been a remarkable lack of agreement on the decay schemes proposed by various authors. It is perhaps worth mentioning some of the experimental problems which hinder a definitive study of these nuclides.

Table 8.7

Beta Transitions of the UX-complex

Nuclide	HEERSHAP[5] Kev	Int.	STOKER[3] Kev	Int.	BRANDT[2] Kev	Int.	ONG[4] Measured Kev	Int.	ONG[4] From Decay Scheme Kev	Int.	JOHANSSON[8] From γ-ray Intensities Kev	Int.	de HAAN[6] Kev	Int.	BJORNHOLM[13] From γ-ray Intensities Kev	Int.
UX_1	192	56%	193	67%	205	80%			193	79%	193	72%	191	65%	.....	.....
	104	44%	103	33%	112	20%			103	21%	101	28%	100	35%		
UX_2	2320	80%	2305	90%					2310	96%	2320	98.8%	2305	96%	2290	98%
	1500	13%	1500	9%					1500	1.0%	1500	0.63%	1500	2.3	2246	~1%
									1300	1.6%	1300	0.49%	1300	1.2	1480	0.72%
	600	7%	580	1%					480	1%	600	0.04%	500	1.4	1245	0.74%
													100	0.5		
	BOUIS-SIERRES[7]															
UZ	1350	7%			1200	10%	1130	13%	1130	16%	1200	10%			1080	7%
	900	18%													730	11%
	500	75%			450	90%	530	27%	530	34%	450	90%				
							320	32%	320	40%					410-520	66%
							155	28%	155	10%					190	14%

Note: SCHNEIDER et al.[14] also report a fermi analysis of the UX_2 spectrum into 11 components; this analysis is inconsistent with the decay scheme described here.

(1) The 1.17 minute UX_2 decays directly to the ground state of U^{234} in 98 percent of its transitions. The continuous beta spectrum accompanying this decay has the high end-point energy of 2300 kev. It is difficult to study the weak conversion electrons in the presence of this beta spectrum. It is particularly difficult to detect the electrons corresponding to the weak (0.13 percent) isomeric transitions to UZ.

(2) UZ has an extremely complex beta, gamma, and conversion electron spectrum which should be studied with spectrometers of high resolution. High resolution implies low transmission however and hence a need for intense sources. But UZ is produced in only 0.13 percent of the decay events of UX_2 which corresponds to a loss in intensity of a factor of 700. To prepare UX_2 sources of millicurie strength one would have to isolate the UX_1 in equilibrium with tons of uranium. The published and unpublished studies cited here were all done with sources of microcurie or smaller intensity so that the necessary high resolution could not be utilized.

A nearly-complete list of references is given at the end of this section of published work on UX_2 and UZ. Work published before 1955 is well reviewed by de HAAN, SIZOO, AND KRAMER.[6] Later detailed discussions are given by ONG, VERSCHOOR, AND BORN,[4] and by SCHNEIDER, DE LANGE, AND DE VILLIERS.[14,15] Rather than attempt a review of these many publications or a resolution of the conflicting interpretations we shall accept the experimental results and interpretation of BJORNHOLM AND NIELSEN[16,17] as representative of a recent comprehensive study of UX_2 and UZ and summarize it briefly here. These authors used the Copenhagen six-gap "orange" type beta spectrometer to measure the electron spectra. They used the same spectrometer in coincidence with a crystal spectrometer to measure beta-gamma, and electron-gamma coincidences. Also they relied on gamma-gamma coincidence and gamma-gamma angular correlation experiments of WOOD.[18]

The beta decay scheme of UX_2 is shown in Figure 8.9. This scheme was constructed in the following way.

The decay energy is set by the end point energy of the intense (98 percent) ground state beta transition. The several determinations of the end point energy listed in Table 8.7 cluster around 2300 kev. The other beta group energies and intensities come chiefly from the decay scheme and gamma intensity analysis. The gamma transitions are listed in Table 8.8.

Table 8.8

Internal Transitions Following the Beta-Decay of UX_2
Bjornholm and Nielsen

Energy kev	Multipolarity	Total Intensity %
43.5	E2	2
K x-ray		
236	E0	0.09
255 ± 5	E1	0.05
746 ± 5	E1	0.04
765	E2	0.30
790 ± 5	E1	0.02
(806) ?	E0	$\leq$ 0.03
811	E0	0.51
1001	E2	0.60
1045	E0	< 0.001
1160		----
1440		~ 0.03
1750		~ 0.03

The 43.5 kev transition is without much question identical with the 43.5 kev transition observed[19] in the alpha decay of Pu^{238} and identified as the de-excitation of the first (2+) level of the ground state rotational band of U^{234}. The 811 kev transition is very strongly converted, as was established by JOHANSSON,[8] and must be electric monopole in character. Consequently there must exist a level at 811 kev with spin and parity 0+. The same 0+ level has been observed by ASARO AND PERLMAN[19] in the alpha decay of Pu^{238} although they assign an energy of 803 kev to this level.

The placement of the other gamma rays in the figure follows chiefly from the electron-gamma and gamma-gamma coincidence results listed in Table 8.9 and from beta-gamma coincidence results listed in Table 8.10 An important fact in the interpretation of the gamma-gamma coincidence data was the recognition that the 236 kev transition is electric monopole in character. This fact also sets the spin and parity of the level at 1045 as 0+. The spin and parity of the level at 790 kev is set at 1- by a measurement of the K-shell conversion coefficient of the 255 kev transition which indicated that it was electric dipole (E1) in character. This assignment was fully confirmed by the experiments of WOOD.[18] He performed gamma-gamma correlation experiments on the 250 - "770" composite cascade in order to provide a crucial check on the assignments of K, I, and π to the levels at 1045 kev and 790 kev. The results confirm the assignments given in the figure. The K, I, π assignments of (0, 1, -) to the level at 790 ± 5 kev is also in agreement with the evidence supplied by GALLAGHER AND THOMAS[20] that such a level appears in the electron capture decay of Np^{234}. GALLAGHER AND THOMAS[20] had intense sources of Np^{234} to work with and were able to set a more precise energy of 788 kev for the energy of this state.

From arguments based chiefly on log ft values one concludes that the most likely ground state spin of Pa^{234} (UX_2) is zero, but 1 is also a possibility.

We reserve comment on the isomeric transition connecting UX_2 and UZ until we have reviewed the decay scheme of UZ.

Many of the authors cited below have contributed valuable measurements of the conversion electrons and gamma photons of UZ but we shall again refer chiefly to the measurements and interpretations of BJORNHOLM AND NIELSEN.[17] These authors measured 51 conversion electron lines which they assigned to the 32 gamma transitions listed in Table 8.11. Several pairs

Table 8.9

Gamma-rays Observed in Coincidence with Conversion Electrons and Gamma-rays in the Decay of UX_2

Selected transition	Gamma-rays found in coincidence with selected transition			Level indicated at:
	Peak energy (kev)	Interpreted as: kev	No. of coincidences	kev
L 43.5	765	746 + 765	18%	811
	1000	1001	32%	1045
K 236	765	765	45%	1045
	(1000)		0%	
255 γ	765	746 + 790		790
770 γ component 746 + 765 + 790	100	K x-rays from 236	~ 12%	
	250	255	~ 10%	
	765	?	≤ 1%	

Results of BJORNHOLM AND NIELSEN[16] and of WOOD.[18]

Table 8.10

Beta-Gamma Coincidence Results on UX_2 Decay

Bjørnholm and Nielsen

End point energy of beta group (kev)	Gamma rays in coincidence		Level Indicated at: kev
	Peak energy kev	Interpreted as	
~ 1500	767	765	811
1250	250	255	1045 + 790
	1000	1001	

Table 8.11

Gamma Transitions in the Decay of UZ

Bjørnholm and Nielsen

Energy (kev)	Multipolarity	K/L		Total Transition Intensity
		Exp.	Theor.	
44	E2	---	---	(92)
100	E2	---	---	71
126	E1			32
153	E2	≤ 0.2	~ 0.2	25
186	M1	5.8	4.4	13
197	M1 or E2	---	---	~ 9
(208)	(E1)			(15)
224	(M1)	4.3	4.4	(7)
228	M1			33
287	E1			(12)
(323)	---	---	---	3
(355)	E1	---	---	(6)
369	M1	4.0	4.5	8.4
565	M1	3.9	4.5	17
694	(M1)	≥ 2.9	4.5	(6)
727	M1	≥ 2.5	4.5	10
791	(E2)	---	---	4
804	(E0)	3.6	---	(0.6)
822	(E2)	---	---	(4)
873				(10)
875	(E2)	4.8	3.5	(5)
878				(12)
920	(E2)	≥ 3.2	3.5	(18)
922				
941	(E2)	~ 4	3.5	13
976	(E2)	---	---	3
1020	(E2)	---	---	(5)
(1130)	---	---	---	---
(1340)	---	---	---	---
(1410)	---	---	---	---
(1620)	---	---	---	3
(1850)	---	---	---	1

of the listed gamma rays have energies so close to each other that their conversion electrons were not in fact resolved from each other. The existence of the pairs (or in some cases of triplets) was deduced from a careful examination of intensity balances and from a detailed consideration of the decay scheme. All these transitions were placed in a tentative self-consistent decay scheme but we reproduce here as Figure 8.9 only that part of it which is established with reasonable certainty; hence only a few of the levels and the gamma transitions are shown. The continuous beta spectrum could not be cleanly separated from the complex conversion electron spectrum which contained 225 conversion electrons for every 100 beta particles. Nonetheless a crude resolution of the beta spectrum into five groups was of considerable assistance in the construction of the decay scheme. It is interesting to note that while the beta decay of UX_2 goes 98 percent to the ground state of U^{234}, the beta decay of UZ goes by several low-energy partial beta groups to excited levels lying between 1000 kev and 2100 kev. A 500 kev beta-group accounts for about 2/3 of the decays. There is no beta-intensity to the ground state rotational band (log ft > 11) which fact may be attributed to K-forbiddeness. A comparison of the decay schemes of UX_2 and UZ suggests a low spin, probably zero (or 1) spin for UX_2 and a high spin, probably 4 for UZ.

The ground state rotational band is represented by the 0+, 2+, 4+, and 6+ levels. There seems little question about the correct assignment of these levels and of the well-characterized E2 cascade radiations by which they are de-excited because the E2 radiations are so intense and because they coincide with those determined in the alpha decay of Pu^{238}. (See Section 9.2.8 of Chapter 9).

The next level of excitation observed in the decay of UZ lies at 922 kev and this is assigned by BJORNHOLM AND NIELSEN[17] to a collective quadrupole vibration of the gamma-vibrational type. Their measurements indicate that no levels appear between 297 kev and 922 kev which is at variance with several decay schemes published by other authors. The first rotational level of excitation of the gamma-vibrational band appears at 965 kev ($K, I, \pi = 2, 2+$). BJORNHOLM AND NIELSEN[17] have tentative evidence for several additional rotational members of the band. Each level must de-excite by E2 transitions to the ground state rotational band. Because the spacings in the two bands are similar many of these transitions are nearly

identical in energy. Hence their conversion electrons were not resolved at the 1.5 percent instrumental resolution which had to be used because of the low intensity of the sources. The nature of the levels at 1150, 1379, 1500, and 1715 kev which are strongly populated by direct beta decay is not established. The spin values are high and the log ft values suggest a spin of 4 with positive parity for UZ. The level at 1374 kev has special interest because of its long half life ($\tau > 2 \times 10^{-7}$ seconds). This delay was discovered when it was noted that the prominent 126 kev E1 transition, present in 32 percent of all disintegrations, was not in coincidence with any radiation within the ground state rotational band. The 126 kev transition is coincident with beta particles. The conclusion is that the level to which the 126 kev transition decays, namely the 1374 kev level, is long lived.

There has been considerable confusion in the literature as to whether UX_2 or UZ is the isomeric state and what the energy of separation is. Most authors conclude that UX_2 is the higher lying of the two. BJORNHOLM AND NIELSEN agree with this choice and set the energy difference as 60 ± 30 kev. If the suggested spins of 0 and 4 for UX_2 and UZ respectively are correct then the two isomers are connected by an M4 or E4 transition.

In the older literature[2,21] the branch decay of UX_2 by isomeric transition to UZ was given as 0.15 percent. In 1954 ZIJP, TOM, AND SIZOO[1] reported the much higher value of 0.63 percent. However a careful restudy of this branching by BJORNHOLM AND NIELSEN[16] reestablished the validity of the earlier determination. They reported 0.13 ± 0.03 percent. They also report evidence for a 73 kev transition in this intensity in the decay of UX_2 which may be the isomeric transition to UZ.

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8.2.13 Thorium-235. Only sketchy information is available on Th^{235}. HARVEY AND PARSONS[1] prepared it by placing a sample of UX_1, which had been separated from one kilogram of uranium, in a flux of 6×10^{13} neutrons. The reactions are:

$$Th^{234}\ (n,\gamma)\ Th^{235};\ Th^{235} \xrightarrow[\text{short}]{\beta^-} Pa^{235} \xrightarrow[23\ \text{min}]{\beta^-} U^{235}$$

Within a few minutes after the end of the irradiation a protactinium fraction was isolated from the thorium and Pa^{235} was identified. No additional Pa^{235} grew into the thorium sample, from which it was concluded that Th^{235} is a beta emitter with a half-life of much less than 5 minutes. The beta decay energy of Th^{235} is estimated as 1.77 Mev from decay cycles.

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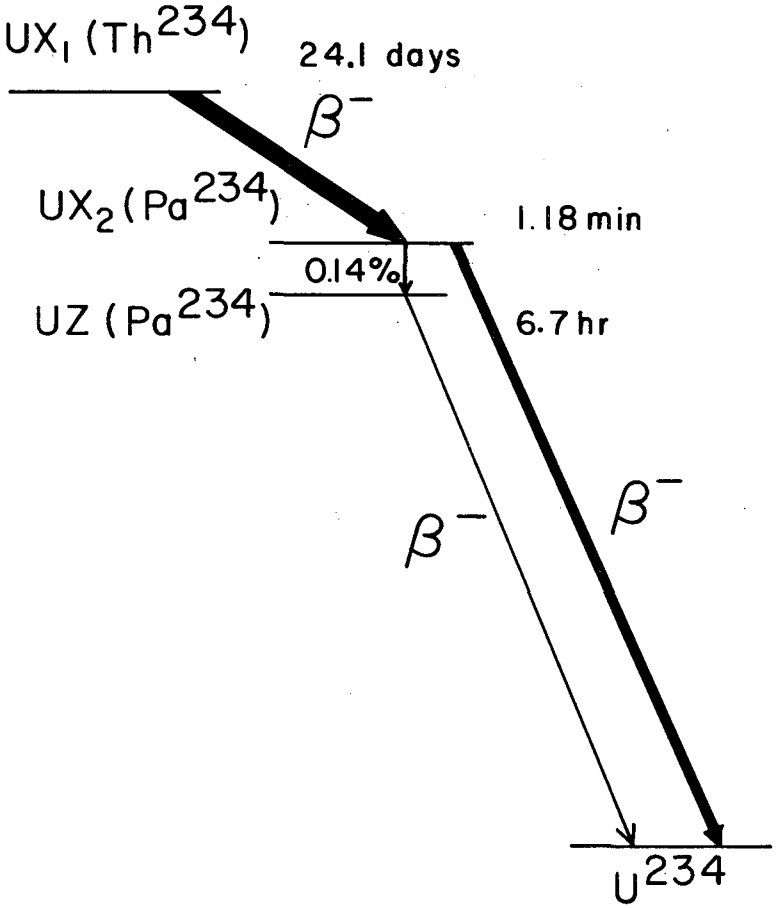
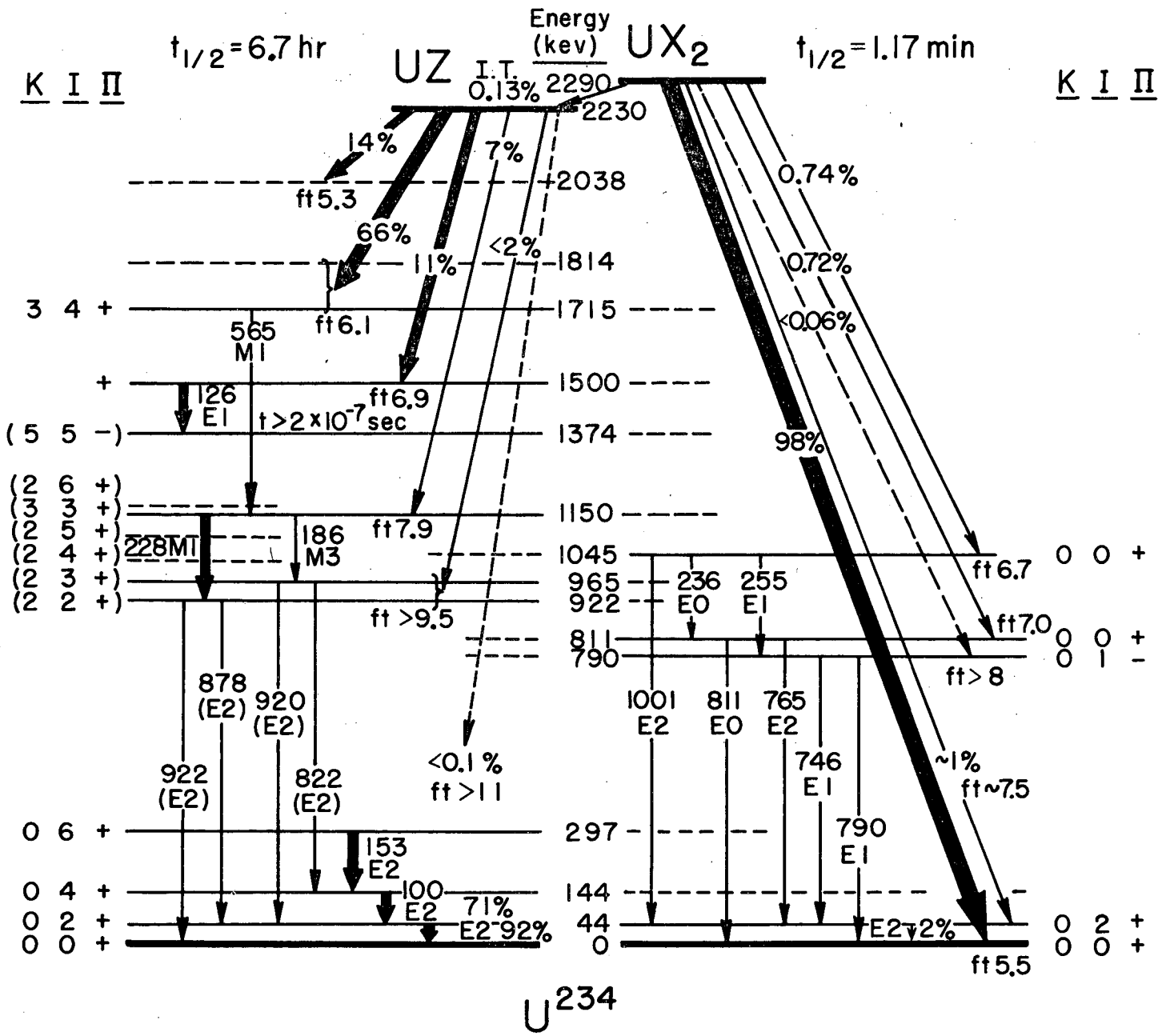


8.8 Genetic relationships in UX-complex. More details of the decay scheme are shown in Figure 8.9.



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8.9 Decay scheme of UX_2 and of UZ as formulated by Bjornholm and Nielsen. Only the most certain features of the schemes are shown.

8.3 THE ISOTOPES OF PROTACTINIUM

8.3.1 Protactinium-225. This isotope is the parent of the Pa^{225} collateral series discussed in Chapter 7. It is believed to be an alpha emitter with a half-life of 2 seconds. The alpha particle energy is estimated as 7.3 Mev. Protactinium-225 is prepared by the bombardment of thorium with high energy protons. The very tentative information on this isotope needs to be confirmed and extended by additional experimental studies.

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8.3.2 Protactinium-226. This isotope is the parent of the Pa^{226} collateral series discussed in Chapter 7. Our total information on Pa^{226} is that it emits alpha particles of 6.81 Mev energy with a half-life of 1.8 minutes.

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8.3.3 Protactinium-227. The discovery of Pa^{227} and its importance as the parent of the Pa^{227} collateral series are discussed in Section 7.2.3 of Chapter 7. Protactinium-227 decays partially (85 percent) by alpha-particle emission and partially (15 percent) by capture of an orbital electron.[1] The resultant half-life is 38.3 ± 0.3 minutes.[1] Orbital electron capture was proved[1] by the isolation of Th^{227} daughter activity.

HILL, ASARO, AND PERLMAN measured the alpha spectrum of Pa^{227} on samples prepared by the bombardment of thorium with 280 Mev protons. Their results are summarized in Table 8.12 and in the figure 8.10.

Because of experimental difficulties the gamma rays accompanying the alpha decay have not been carefully studied so that it is not possible to draw a detailed decay scheme. HILL interprets the Ac^{223} level at 67.3 kev as a K = 5/2 state of intrinsic excitation upon which a series of rotational levels is based. As indicated in the figure the levels at 109.7, 157.9, and 208.6 kev may be the 7/2, 9/2, and 11/2 levels of this rotational band. A suitable NILSSON orbital assignment would be 5/2- [523]. Since the favored alpha decay of Pa^{227} decays to this level the suggested assignment for the ground state of Pa^{227} is also 5/2- [523]. This is also the favored assignment for Pa^{229} whose alpha decay scheme is similar to that of Pa^{227}. These details of interpretation are tentative and other major features of the decay of Pa^{227} are unknown both experimentally and theoretically.

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1. W. W. Meinke, A. Ghiorso, and G. T. Seaborg, Phys. Rev. 81, 782 (1951).

2. M. W. Hill, F. Asaro, and I. Perlman, unpublished results, 1957.

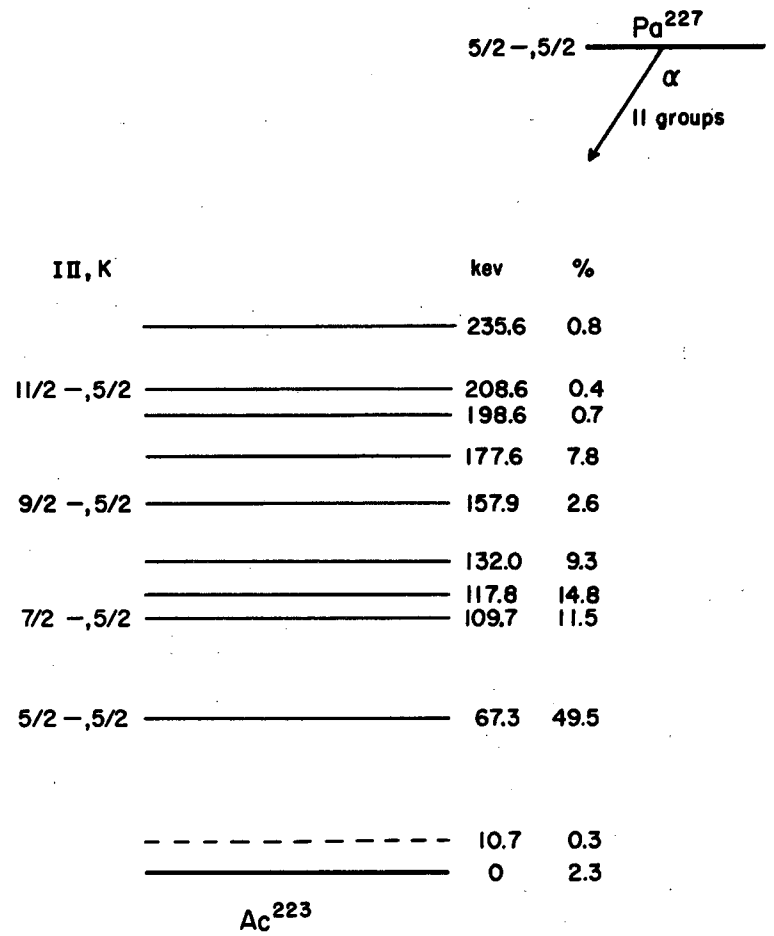
3. M. W. Hill, Thesis, University of California, 1958; See University of California Radiation Laboratory Report UCRL-8423.

Table 8.12

Pa^{227} Alpha Groups

Alpha-particle energy (Mev)[a]	Excited-state energy (kev)	Abundance (%)	Hindrance factor
6.526	0	2.3	130
6.515	10.7	0.3	900
6.460	67.3	49.5	3.1
6.418	109.7	11.5	8.8
6.410	117.8	14.8	6.3
6.396	132.0	9.3	8.8
6.371	157.9	2.6	24
6.351	177.6	7.8	6.6
6.331	198.6	0.7	60
6.321	208.6	0.4	95
6.294	235.6	0.8	36

[a] Relative to $Bi^{211}_{\alpha_{354}}$ = 6.273 Mev.



8.10 Decay scheme for the alpha decay of Pa^{237}.

8.3.4 Protactinium-228. The discovery of the 22 hour Pa^{228} and its position as the precursor of the Pa^{228} series of radioactive isotopes is discussed in Section 7.2.2 of Chapter 7. Protactinium-228 is prepared by the bombardment of thorium with high energy protons or deuterons and all samples are necessarily contaminated with some of the higher-mass protactinium isotopes. This interferes with the careful study of its complex radiations particularly its gamma radiations. This contamination can be minimized by proper selection of the energy of the bombarding particles; for protons an energy of 65 Mev is best. The contamination can also be eliminated entirely by electromagnetic separation of Pa^{228} in a suitable isotope separator. Protactinium-228 disintegrates 98 percent by capture of an orbital electron and two percent by alpha decay.

HILL, ASARO, AND PERLMAN[1,2] have studied the alpha spectrum of Pa^{228} in a double-focusing magnetic spectrometer of high resolution and have found it to be exceedingly complex. Table 8.13 lists twenty-seven alpha groups. The Ac^{224} levels revealed by these alpha groups are also listed in the table and shown in Figure 8.11. No interpretation of these levels was advanced by these authors.

Obviously the gamma transitions resulting from the de-excitation of these many excited levels must be numerous. HILL[2] measured the gamma spectrum in coincidence with Pa^{228} alpha particles. A complex spectrum was noted in the energy region 90 to 400 kev which was roughly resolved into the components listed in Table 8.14. These gamma rays were not placed in the decay scheme.

The electron capture decay of Pa^{228} have been studied by several authors. ONG[3,4] and HILL[1,2] and HILL, HOLLANDER, AND PERLMAN[5] measured conversion electron lines in magnetic spectrometers and gamma ray photons in sodium iodide crystals. From intensity, multipolarity and coincidence measurements these authors were able to construct partial decay schemes and to interpret the low-lying levels in the daughter nucleus Th^{228}. A later and more complete study was made by ARBMAN, BJØRNHOLM, AND NIELSEN.[6] We quote here exclusively from this study.

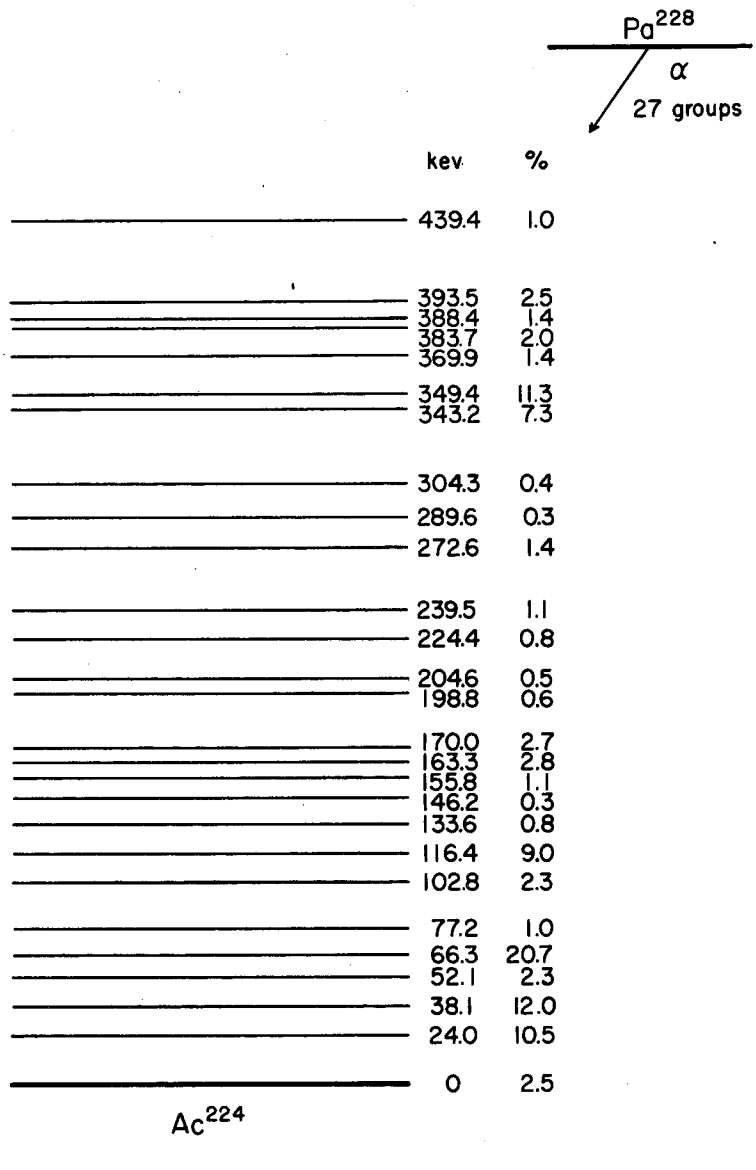
These authors studied the conversion electrons in an 18.5 cm double focusing electron spectrometer adjusted to a resolution of 0.2 percent and a transmission of 0.2 percent. They also used a six-gap, orange type spectrometer adjusted to a resolution of 0.4 percent and a transmission of

Table 8.13

Alpha groups of Pa^{228}

Alpha-particle energy (Mev)	Excited-state energy (kev)	Abundance (%)	Hindrance factor
6.138	0	2.5	3200
6.114	24.0	10.5	580
6.101	38.1	12.0	350
6.087	52.1	2.3	2000
6.074	66.3	20.7	190
6.062	77.2	1.0	3400
6.037	102.8	2.3	1100
6.024	116.4	9.0	260
6.007	133.6	0.8	2300
5.994	146.2	0.3	5400
5.985	155.8	1.1	1300
5.978	163.3	2.8	480
5.971	170.0	2.7	460
5.943	198.8	0.6	1500
5.937	204.6	0.5	1700
5.918	224.4	0.8	850
5.903	239.5	1.1	520
5.870	272.6	1.4	280
5.854	289.6	0.3	1100
5.839	304.3	0.4	610
5.801	343.2	7.3	24
5.795	349.4	11.3	14
5.775	369.9	1.4	92
5.761	383.7	2.0	55
5.756	388.4	1.4	73
5.752	393.5	2.5	43
5.707	439.4	1.0	57

These energies were measured relative to $U^{230}_{\alpha_o}$ = 5.884. From thesis of HILL.



8.11 Alpha decay scheme of Pa^{228}.

Table 8.14

Gamma rays in Pa^{228} alpha decay

as roughly resolved by HILL

from gamma spectrum in coincidence with alpha particles

E_γ (kev)	Relative intensity	E_γ (kev)	Relative intensity
95	240	220	10
130	27	240	55
150	34	280	49
170	11	310	100
200	14	345	21

one percent. The gamma ray transition energies deduced from these measurements are listed in Table 8.15. One hundred fifty three conversion lines from 72 gamma transitions were observed. The transitions below 500 kev were characterized in multipolarity by their $L_I + L_{II}/L_{III}$ and K/L electron ratios. Some information on photon intensities was obtained from a NaI scintillation spectrometer. Very important information on the decay scheme was obtained by electron-gamma coincidence experiments in which the coincidence circuit was gated by the detection of electron lines of a specific energy (resolution 1.5 percent) and the gamma spectrum coincidence was measured with a multichannel scintillation spectrometer. About 50 coincidence spectra were recorded involving the conversion lines of 32 transitions and the photo peaks of 50 different gamma-rays. These measurements were aided by the fact that there was no beta spectrum to provide an interfering background.

From a detailed consideration of all the evidence ARBMAN, BJØRNHOLM, AND NIELSEN[6] constructed the very detailed decay scheme shown in Figure 8.12. This scheme has 22 levels ranging in energy from 57.5 kev to 1943 kev. Considerable help in interpreting the data relating to the lower levels was obtained from a consideration of the alpha decay of U^{232} and the beta decay of Ac^{228} to the same Th^{228} daughter nucleus. For a discussion of these nuclides see Sections 8.4.6 and 6.3, respectively. Figure 8.12 accounts for 47 of the 72 transitions found. The intensity of this fraction of the transitions amounts to 310 per 100 electron capture decays, while the 25 transitions not included represent 30 out of 100 transitions. Most of the levels shown in the scheme were cross checked in many ways and are firmly established. The intensity balance for the flow of transitions through the excited states to the ground state comes out in a consistent way. Table 8.16 gives a list of the levels and the intensity of feeding and de-excitation of each level. By difference the electron capture intensity and log ft values were calculated.

The level system of Th^{228} has been interpreted in the language of the collective model of the nucleus. It is one of the best examples in the whole heavy element region of an even-even nucleus with many levels which can be correlated with the rotational and vibrational excitations predicted by the theory. We now mention several of these features.

Table 8.15

Gamma transitions in the electron capture decay of Pa^{228}

Arbman, Bjørnholm, and Nielsen[6]

Energy (kev)	Multipolarity	Total intensity	Placed in decay scheme
57.5	E2	82	+
99.4	M1	0.7	+
129.1	E2	21	+
137.8	M1	2.0	
178.0	M1	0.85	
184.5	M1	2.2	+
(191.3)	(E2)	~1	(+)
209.0	E1	3.8	+
224.0	M1	3.6	+
270.0	E1	3.2	
(278)	(E2)	(0.9)	
282.2	(M1 + E2)	4.0	+
327.5	E1	3.3	+
327.5	[E2]	2.5	+
327.5	E1	3.3	+
338.5	E1	7.8	+
341.1	E2	2.8	+
(397)			
409.7	E2	14	+
(462)	(E2)	3	+
463.3	E2	26	+
(469.2)			
(617)			
(622)			
641			
662			
666			
(670)			
680			
694			

(continued)

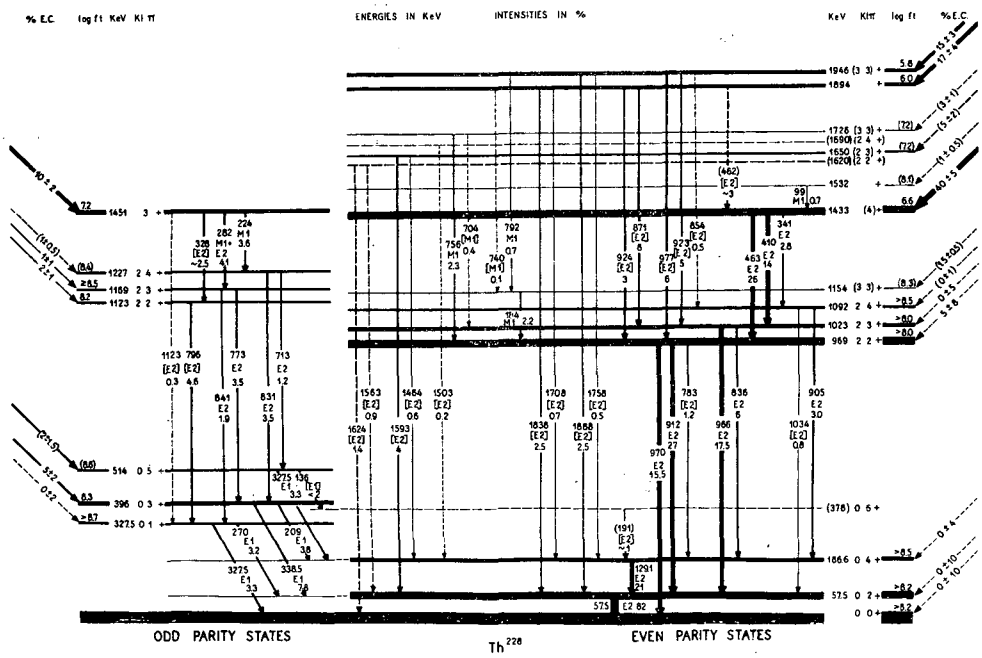
Table 8.15 - continued

Energy (kev)	Multipolarity	Total intensity	Placed in decay scheme
704	[M1]	0.4	(+)
713	E2	1.2	+
732			
739.6	[M1]	0.13	(+)
745.6			
756.2	M1	2.3	+
773.4	E2	3.5	+
782.6	[E2]	1.2	+
792.2	M1	0.7	+
795.8	[E2]	4.6	+
817.4			
831.4	E2	3.5	+
835.8			
836.4	E2	6.0	+
841.0	E2	1.9	+
853.8	[E2]	0.5	(+)
871.0	(E2)	8.0	+
889.0			
905.2	E2	3.0	+
912.2	E2	27	+
923	(E2)	5	+
924	(E2)	3	+
966	E2	17.5	+
970	E2	15.5	+
976.8	E2	6	+
1034.1	(E2)	0.8	+
(1123)	(E2)	0.3	(+)
1168			
1253	(M1)		
(1293)			
(1423)			
(1464	(E2)	0.6	+
(1489)			

(continued)

Table 8.15 - continued

Energy (kev)	Multipolarity	Total intensity	Placed in decay scheme
(1503)	(E2)	0.2	+
1563	(E2)	0.9	+
1593	(E2)	4	+
1624	(E2)	1.4	(+)
1678			
1708	(E2)	0.7	+
1744	(E2)		
1758	(E2)	0.5	+
1838	(E2)	2.5	+
1888	(E2)	2.5	+



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8.12 The decay scheme of Pa^{228} for capture of an orbital electron as formulated by Arbman, Bjørnholm, and Nielsen. The thickness of the lines reflects roughly the intensity of the transitions or the extent to which each level is populated. The odd parity levels have been separated from those of even parity. The electron capture transitions are therefore seen to come in from both sides. Transitions and levels for which only tentative evidence is obtained are dotted. Similarly numbers in parentheses are tentative.

Table 8.16

Levels in Th^{228}

Total intensities of internal transitions feeding and de-exciting each level

Electron-capture intensities and log ft-values

Pa^{228} : $t_{1/2}$ = 22 ± 1h Q_{oEC} = 2.10 ± 0.05 Mev

		Pa^{228}				Ac^{228} (a)
Level kev	K,I,π	Internal transitions: feeding level %	Internal transitions: de-exciting level %	EC directly feeding level %	log ft	log ft
0	0,0 +	101	0	0 ± 10	> 8.2	> 8.8
57.5	0,2 +	86	82	0 ± 10	> 8.2	8.8
186.6	0,4 +	20	21	0 ± 4	> 8.5	-
327.5	0,1 +	6.8	6.5	0 ± 2	> 8.7	> 9.5
395.8	0,3 +	7.0	11.6	5 ± 2	8.3 ± 0.3	8.5
514.1	0,5 +	1.2	3.3	(2 ± 1.5)	(8.6 ± 0.4)	-
969.5	2,2 +	39.5	43.7	5 ± 8	≥ 8.0	7.3
1023.2	2,3 +	27.5	23.5	0 ± 5	> 8.0	≥ 8.0
1091.8	2,4 +	3.3	3.8	0 ± 1	> 8.5	-
1123.4	2,2 +	2.5	4.6	2 ± 1	8.2 ± 0.3	8.0
1168.8	2,3 +	4.0	5.4	1 ± 1	> 8.5	-
1227.0	2,4 +	(3.6)	4.7	(1 ± 0.5)	(8.4 ± 0.3)	-
1154.0	(3,3)+	(0.8)	2.2	(1.5 ± 0.5)	(8.3 ± 0.3)	7.7
1432.8	(4)+	3.7	42.8	40 ± 5	6.6 ± 0.1	> 8.0
1451.0	(3)+	0	10.1	10 ± 2	7.2 ± 0.1	≥ 8.3

(continued)

Table 8.16 continued

Level kev	K,I,π	Pa^{228} Internal transitions: feeding level %	de-exciting level %	EC directly feeding level %	log ft	Ac^{228} (a) log ft
1532.2	(4)+	0	0.7	(1 ± 0.5)	(8.1 ± 0.4)	7.1
(1620)	(2,2)+	(0)	(2.3)	(2 ± 1)	(7.6 ± 0.5)	-
1650	(2,3)+	0	4.6	(3 ± 2)	(7.2 ± 0.3)	6.8
(1690)	(2,4)+	(0)	(0.2)	(0.2 ± 0.1)	(8.5 ± 0.5)	-
1726	(3,3)+	0	2.7	(3 ± 1)	(7.2 ± 0.3)	-
1894.0	+	0	17	17 ± 4	6.0 ± 0.4	-
1946.3	(3,3)+	0	15	15 ± 3	5.8 ± 0.4	-
Total		206	307	111 ± 20		

(a) Reference 6 and unpublished results of the authors of the same reference.

The level structure clearly shows the presence of rotational bands. built on the ground state (0+), the 327.5 kev level (1-), the 969 kev level (2+), the 1123 kev level (2-), and possibly on a 1620 kev level (2+). Three members of each band are excited and it is possible to fit them all to the standard rotational formula

$$E_{rot} = E_o + \frac{\hbar^2}{2\Im} I(I+1) + B I^2 (I+1)^2$$

Where I is the spin, $\hbar$ is plancks constant divided by 2π, $\Im$ is the effective moment of inertia and B is a constant giving the magnitude of a small second order correction for the effects of particle-rotation interaction. See Chapter 3 for a more complete discussion of this formula. The constants of this equation are fitted to the 5 observed rotational bands as shown in Table 8.17.

The ground state rotational band is very plainly seen in the decay of Ac^{228} and U^{232} and resembles that observed in all neighboring even-even nuclei; i.e. it has the level sequence 0+, 2+, 4+ ··· and the higher levels are de-excited by E2 transitions to the level below. The K-quantum number for the band is 0. BELL, BJØRNHOLM, AND SEVERIENS[7] have measured the life-time of the first excited 57.5 kev state by a delayed coincidence technique and have found a value $t_{1/2} = 4.0 \times 10^{-10}$ seconds. From this they calculated an intrinsic electric quadrupole moment, $Q_o = (8.5 \pm 0.3) \times 10^{-24}$ cm^2. This transition is enhanced by a factor of 200 compared to the single particle estimate which agrees with the prediction of the collective model.

The even parity band with the 969 kev 2+ base state is assigned to a quadrupole gamma-vibration of the nuclear shape. The K-value for this band is 2. The level sequence of the 969, 1023, 1092 kev levels has the spacing, the 2+, 3+, 4+ sequence, and the gamma-ray de-excitation pattern expected for the gamma-vibrational band. See the discussion of this point in Chapter 3. The de-excitation occurs by E2 transitions to the ground state rotational band. When 2 or more of these electric quadrupole transitions originate in one state and terminate in different members of the ground state rotational band the relative intensities according to theory are given by simple geometrical factors (Clebsch-Gordan coefficients) after the energy dependence is factored out. Several comparisons of experimental with theoretical ratios were carried out by ARBMAN, BJØRNHOLM, AND NIELSEN[6] for transitions originating in the gamma-band and in the other bands.

Table 8.17

Rotational Bands in Th^{228}

$$E_{rot} = E_o + A\,I\,(I+1) + B\,I^2\,(I+1)^2$$

Band Head		$A = \hbar^2/2\,\Im$	- B
kev	K I π	(kev)	(kev)
0	0 0+	9.70 ± 0.03	0.020 ± 0.002
327.5	0 1-	6.96 ± 0.03	0.009 ± 0.001
969	2 2+	9.43 ± 0.07	0.027 ± 0.003
1123	2 2-	7.93 ± 0.08	0.021 ± 0.004
(1620)	(2 2+)	(5.0 ± 0.5)	

Arbman, Bjørnholm and Nielsen

In addition to the quadrupole collective nuclear vibrations, the collective model predicts the probable occurrence of octupole vibrations having negative parity and K = 0, 1, 2, or 3. The negative parity state at 327.5 kev is believed to be the K = 0 octupole vibration. It is observed in the alpha decay of U^{232} (See Section 8.3.5). It decays by a pair of E1 transitions to the 0+ and 2+ levels of the ground state band and hence is well characterized as a 1- state. The rotational levels based on this 1- state should, according to theory, have spin and parity values 1-, 3-, 5-, etc. and an energy spacing given by the I (I + 1) rule. The levels at 396 and 514 kev are identified as the predicted 3- and 5- levels.

The 2-, 3-, 4-, rotational series based on the 2+ level at 1123 kev may represent a K = 2 octupole vibration. However the evidence for this is not clear and it has also been suggested that this group of levels represents a gamma-vibration superimposed on the K = 0 octupole band.[6]

Some speculative suggestions have been made concerning the collective nature of some other levels in the decay scheme but these are not recorded here.

A consideration of the decay scheme and the spin values, parities, and log ft values of Table 8.16 leads to the assignment of spin 3 and positive parity to Pa^{228}. One can make a logical correlation of this assignment with the Nilsson diagrams for odd neutron and proton states in the following way. From the Nilsson diagrams given in Chapter 3 one finds for the 137th neutron the orbit 5/2- [752] and for the 91st proton the orbit 1/2- [530]. The deformation parameter beta corresponding to these choices is 0.225. The spin-coupling rule for odd-odd nuclei formulated by GALLAGHER AND MOSZKOWSKI,[8] leads to the desired value 3+.

The decay scheme of Ac^{228} has an overall similarity to that of Pa^{228} which is explained by the fact that Ac^{228} also probably has spin 3 with positive parity.[6]

The applicability of the collective model to the description of the Pa^{228} decay scheme is also shown in the operation of the K-selection rules. It is clear from a short examination of the scheme that some selection rules other than the usual ones depending on total spin and parity must be in operation. For example those states having quantum number K = 0 are

poorly populated or missed entirely in the direct electron decay process. One striking feature is the complete absence of the expected beta-vibrational band with K = 0 which would be expected at approximately 1 Mev of excitation. In addition the K-selection rule serves to explain why the high energy states decay chiefly by multiple cascade instead of decaying directly to the appropriate levels in the ground state band.

For further details on the decay of Pa^{228} the references cited, particularly the paper by ARBMAN, BJØRNHOLM, AND NIELSEN[6] should be consulted.

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1. M. W. Hill, F. Asaro, and I. Perlman, unpublished information, 1957.
2. M. W. Hill, thesis, University of California 1958; University of California Radiation Laboratory Report UCRL-8423, August 1958.
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8.3.5 Protactinium-229. This isotope was first produced[1] by the bombardment of ionium with deuterons which gives rise to 1.4-day Pa^{229} by the reaction:

$$Th^{230}\ (d,3n)\ Pa^{229}.$$

The Pa^{231} prepared simultaneously by the (d,n) reaction does not interfere in radioactivity measurements because of its low specific activity. There is more interference from 17-day Pa^{230} prepared by the (d,2n) reaction. If the ionium contains a large isotopic impurity of Th^{232}, as it usually does, then Pa^{232} and Pa^{233} radioactivity is also produced and interferes with a detailed study of Pa^{229}.

Protactinium-229 decays almost entirely by electron capture, and has an alpha-branching of only 0.25 percent.[2] In spite of its small alpha-branching, the proof of the mass assignment of Pa^{229} was made by the identification of its Ac^{225} and Fr^{221} daughter activities.[1]

The electron-capture decay of Pa^{229} has been studied by ONG[3] and by HILL, HOLLANDER, AND PERLMAN[4] who recorded the conversion electrons on photographic emulsions placed in permanent magnet spectrometers. ONG found L_{II} and L_{III} conversion lines of a 41.7 kev transition, while HILL AND CO-WORKERS found L_I, L_{II}, L_{III}, M_I, M_{II}, M_{III}, N_{II}, N_{III} lines corresponding to a transition of 42.37 kev. The L_I/L_{II} and L_{III}/L_{II} ratios indicate that the multipolarity of the transition is 95% E2 - 5% M1. This is in good agreement with the results of alpha-decay studies of U^{233}, [5] which define levels in Th^{229} at 0, 42.8, and 98.9 kev as being members of a rotational band with spins of 5/2, 7/2, and 9/2.

The fact that the electron-capture decay of Pa^{229} populates the I = 7/2 member of the rotational band is consistent with the assignment of spin 5/2 to Pa^{229} rather than 3/2 as is the case in Pa^{231} and Pa^{233}. The Nilsson level assignment of 5/2- [523] has been suggested for the ground state of Pa^{229} where the quantum numbers in brackets are [N, N_z Λ].

The alpha spectrum of Pa^{229} has been investigated by HILL[6,7] in a high-resolution, double-focussing magnetic spectrometer. He found a very complex spectrum with the groups listed in the Table 8.18 and shown in the Figure 8.13. HILL[7] performed alpha-gamma coincidence experiments to determine what gamma rays occurred in the alpha-branching decay of Pa^{229}. The gamma rays detected had the following energies and abundances per alpha transition:

Table 8.18

Alpha Groups of Pa^{229}

Alpha-particle energy (Mev)	Excited-state energy (kev)	Abundance (%)	Hindrance factor
5.665	0.0	19.1	23.
5.625	40.5	9.8	28.
5.610	56.3	13.4	170.
5.586	80.6	4.7	36.
5.575	91.9	36.8	4.0
5.560	107.3	3.9	31.
5.531	136.7	8.9	9.4
5.512	155.9	0.60	110.
5.496	172.1	0.74	73.
5.474	194.1	1.77	23.
5.417	252.4	0.07	280.
5.408	263.7	0.15	115.
5.315	356.0	0.05	100.

Standards used in setting energy scale -- $U^{230}_{\alpha_o}$ = 5.884 Mev and $Ra^{224}_{\alpha_o}$ = 5.681 Mev.

The hindrance factor is the factor by which the observed partial alpha half-life differs from that calculated by simple barrier penetration theory.

40 kev (0.10), 69 kev (0.05), 81 kev (0.02), 92 kev (0.16), 107 kev (0.05), and 120 kev (0.02). From crude estimates of the upper limits to the conversion coefficients for these gamma rays it is certain that the prominent 40 and 92 kev transitions are electric dipole (E1) in nature and probable that the others also are E1. Only the 40 and 92 kev transitions are placed in the decay scheme. From the complexity of the alpha spectrum it is [illegible] certain that a larger number of gamma rays accompany the alpha decay of Pa^{229} than have been identified so far, but the experimental difficulties in the way of a careful study of these low intensity transitions are large.

It is likely that the numerous observed levels of Ac^{225} can be accounted for by rotational bands of levels based on only a few states of intrinsic excitation of nucleonic motion. HILL[7] has attempted such an analysis. He concludes that the favored alpha decay proceeds to the 91.9 kev level in Ac^{225} to which he assigns the Nilsson level 5/2- [523]. The rotational band based on this intrinsic state gives rise to a 7/2- level at 136.7 kev, a 9/2- level at 194.1 kev and an 11/2- level at 263.7 kev. The level at 56.3 kev he assigns K-quantum number 5/2- and parity positive and concludes that rotational levels with character 7/2+, 9/2+, and 11/2+ occur at 107.3 kev, 172.1 kev, and 252.4 kev. The ground state of Ac^{225} is believed to have spin 3/2 and probably has the same Nilsson assignment as does Ac^{227}, namely 3/2+ [651]. HILL relied on the energy spacing formula for rotational bands in odd-A nuclei as discussed in Chapter 3 and the considerations of BOHR, FROMAN, AND MOTTELSON[8] on the pattern of alpha abundances to various members of a rotational band.

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2. L. M. Slater and G. T. Seaborg, unpublished data (Aug. 1951).

3. Ong Ping Hok, thesis, Free University of Amsterdam (March, 1955).

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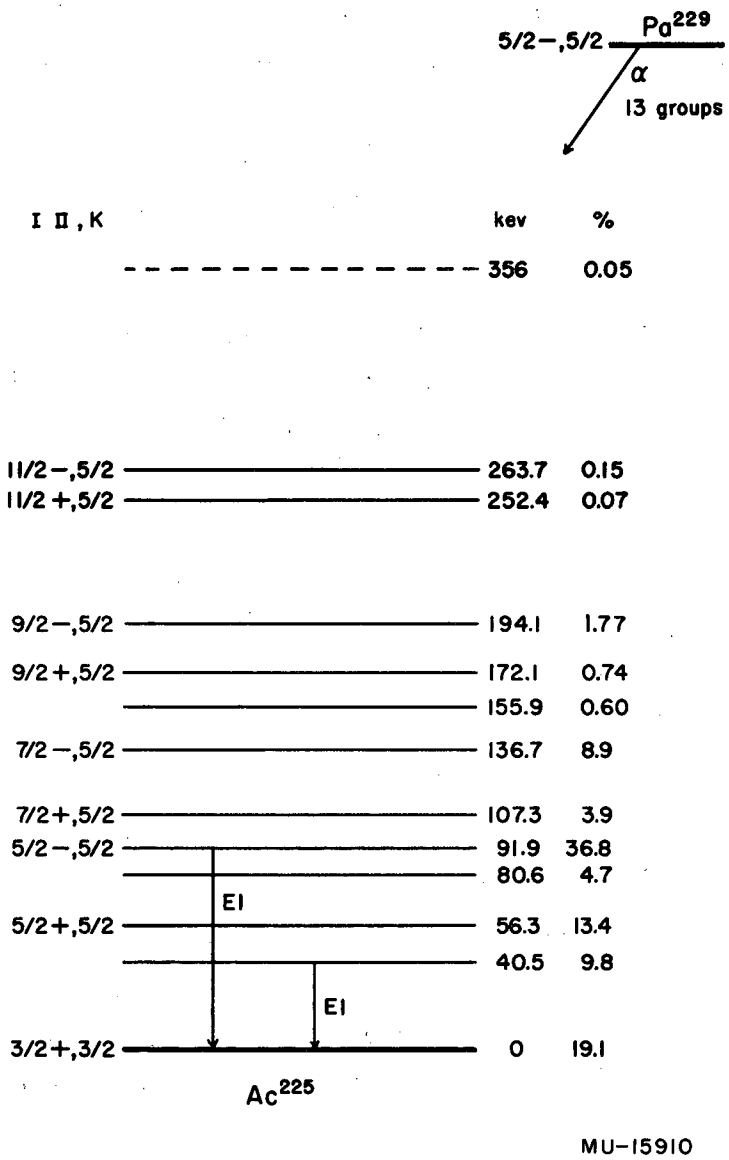
8. A. Bohr, P. O. Froman, and B. R. Mottelson, Kgl. Danske Videnskab Selskab. Mat. Fys. Medd. 29, No. 10 (1955).



8.13 Decay scheme for the alpha decay of Pa^{229} as given by Hill.[7] The spin, parity, and K quantum number assignments on the left of each level are speculative.

8.3.6 Protactinium-230. This isotope is the parent activity of the Pa^{230} collateral series. The discovery of Pa^{230} by STUDIER AND HYDE[1] and its relationship to the other members of the series are discussed in Section 7.2.1 of Chapter 7.

Samples of Pa^{230} can be made by bombardment of thorium with protons. The energy should be chosen so as to emphasize the reaction,

$$Th^{232}\ (p,3n)\ Pa^{230}$$

TEWES[2] reports an excitation function with a peak yield of 400 millibarns at a proton energy of 24 Mev. Some production of Pa^{232} by the (p,n) reaction cannot be avoided but this isotope decays out more rapidly than Pa^{230}. A smaller amount of Pa^{233} produced by the (p,γ) reaction may be more troublesome because of the similarity in half-lives.

Protactinium-230 can also be produced by the reactions:

$$Th^{232}\ (d,4n)\ Pa^{230}$$
$$Io^{230}\ (d,2n)\ Pa^{230}$$
$$Io^{230}\ (p,n)\ Pa^{230}$$
$$Pa^{231}\ (n,2n)\ Pa^{230}$$
$$Pa^{231}(d,p2n)\ Pa^{230}$$
$$Pa^{231}(p,pn)\ Pa^{230}$$
$$Th^{232}\ (\alpha,p5n)Pa^{230}$$

The half life of Pa^{230} has been reported as 17.0 ± 0.5 days[1] and 17.7 ± 0.5 days.[3] The radioactive decay of Pa^{230} proceeds by three distinct modes. The most readily apparent is that of beta emission because the alpha radiation of the daughter resulting from the beta-decay mode is so prominent. In fact, however, only 10 percent of the decay goes by this mode. Approximately 90 percent of the decay is by electron capture to produce ionium, a long-lived product (STUDIER AND BRUEHLMAN[4]). In addition, MEINKE AND SEABORG[5] have shown that in ~0.003 percent of the disintegrations, an alpha particle is emitted and Ac^{226} is produced. It is possible that Pa^{230} undergoes positron emission to an extent somewhat less than 0.1 percent.[6,7]

The numerous gamma radiations emitted in the electron capture branch have been studied by ONG[6,7], by HILL[8] and by NIELSEN AND CO-WORKERS.[9] Since the work of the last-mentioned group is the most thorough we shall quote almost exclusively from it.

These workers measured the conversion electron spectrum carefully in an iron-free six gap beta spectrometer adjusted to a resolution of 0.5 percent. The photon spectrum was measured with a sodium iodide scintillation spectrometer. Coincidence experiments were performed in which the coincidence circuit was triggered by pulses from the beta spectrometer registering a specific conversion line and the entire coincident photon spectrum was measured with a sodium iodide spectrometer. These experiments were very useful in the construction and checking of the decay scheme and were carried out for every principal gamma transition. In addition some electron-electron coincidences were studied.

Table 8.19 lists the gamma transitions together with their multipolarities and their total intensity. The multipolarity assignments were made by a comparison of experimental conversion coefficients with theoretical values. The level scheme of Th^{230} which is deduced from this work is shown in Figure 8.14. This nucleus is a fine example of the experimental confirmation of the collective excitations predicted by the unified model of the nucleus for even-even nuclei.

The lowest levels are the 0+, 2+, 4+, members of the ground state rotational band observed universally for even-even nuclei in the mass region above mass ~226. The typical cascade of E2 transitions is observed. HILL, HOLLANDER, AND PERLMAN[10] thoroughly characterized these E2 transitions and measured precise values of 53.15 kev and 120.8 kev for their energy. ONG[11] called attention to the presence of the ground state rotational band in Th^{230}. The next group of levels is a negative parity rotational band consisting of the two states at 508 and 573 kev. The spin and parity of these are firmly fixed as 1- and 3-, respectively, by their pattern of de-excitation via pairs of electric monopole transitions to the ground state rotational band. The K-quantum number of this band was found to be 0 from the ratio of the reduced transition probabilities of the two E1 transitions de-exciting these levels. According to theory this ratio is simply the ratio of squares of Clebsch-Gordan coefficients. This negative parity, K = 0 band is believed to represent an octupole vibration of the nuclear shape. A negative parity rotational band of this nature is a general feature of neighboring even-even nuclei.

Another generally occurring collective vibration is the so-called beta-vibration of the quadrupole type. In the Th^{230} nucleus the 0+ level

at 634 kev is of this type. The spin and parity of this level is fixed at 0+ by the observation of conversion lines of an E0 transition of 634 kev energy and the determination that these electrons are not in coincidence with electrons of the 53 kev transition in the ground state rotational band. The E0 nature of the transition was confirmed by the establishment of a lower limit of ~ 7 for the K-conversion coefficient. The first rotational excitation of the beta-vibrational band occurs 42 kev higher at 676 kev where a 2+ level exists. This level was identified by observing electrons of a 623 kev transition in such abundance that the multipolarity could only be electric monopole and by observing that these electrons were in coincidence with the electrons of the 53 kev transition in the ground state rotational band. This level was also identified independently in quite a different way by CLASS and his CO-WORKERS[12] at Rice Institute. By bombardment of ionium targets with low energy protons these experimentalists coulombically excited this 676 kev level. They observed the conversion electrons of the 623 E0 transition.

The gamma-mode of the quadrupole vibrations may be identified with the 2+ level at 783 kev. The placement of this state is confirmed by two E2 transitions of 730 kev and 783 kev energy, the first of which is in coincidence with the 53 kev transition in the ground state band.

The level at 954 kev is very important in the decay of Pa^{230} as it is this level which is most strongly populated in the beta decay. This level is strongly de-excited by a 954 kev electric dipole transition going directly to the ground state which fixes the spin and parity of the 954 kev state as 1-. This transition is seen in about half the disintegrations of Pa^{230}. The 2- level at 974 may be a rotational excitation of the 954 kev level. There is some unpublished speculation that this band may represent a mode of octupole vibration.

The decay scheme shows a number of dotted transitions with energy 280, 337, 320, and 380 kev which are not observed in the conversion electron or singles gamma spectrum. The photons of gamma rays of approximately this energy are plainly seen in the gamma spectrum coincident with the conversion electrons of the two E0 transitions.

The decay scheme leads to the selection of 2- as the most likely spin and parity for Pa^{230}. One can account for this choice by selecting the Nilsson orbit 1/2- [530↑] for the 91st proton (by analogy to the 91st proton in Pa^{231}) and the orbit 5/2+ [633↓] for the 139th neutron (by analogy

to the 139th neutron in Th^{229}). By the GALLAGHER-MOSZKOWSKI[13] coupling rule these choices lead to the resultant spin and parity, 2-, for the ground state. The Nilsson assignments cannot be considered proved on this evidence alone.

The absence of any direct beta population to the lower levels of ionium below 700 kev is explained by the K-selection rule.

Very little is known about the beta decay branch of Pa^{230}. ONG[7,8] has reported an end-point energy of 410 kilovolts for the beta spectrum. HILL[8] has measured conversion electrons of a 51.67 kev transition with the L_{II}, L_{III}, M_{II}, and M_{III} spacings corresponding to conversion in uranium rather than thorium. Hence this transition occurs in U^{230} following beta decay and reveals the 2+ first rotational excitation to the ground state.

Table 8.19

Gamma Transitions in the Decay of Pa^{230}

To be completed

Table 8.20

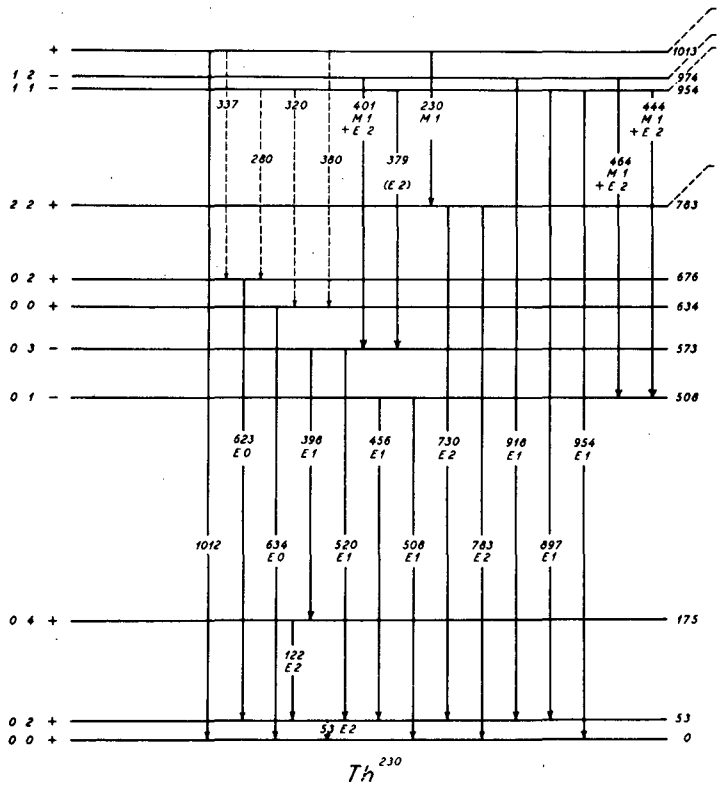
Fitting of Rotational Bands of Ionium
to the formula $E = E_o + A\,I\,(I + 1) + B\,I^2\,(I + 2)^2$

To be completed

Table 8.21

Evaluation of K-Quantum Numbers from Clebsch-Gordan Coefficient Ratios

To be completed



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8.14 The level scheme of Th^{230} and the pattern of gamma ray de-excitation observed in the orbital electron capture of Pa^{230}. All energies are given in kev. The 954 kev level is most strongly populated. Figure obtained from O. B. Nielsen.

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11. Ong Ping Hok, Phys. Rev. 99, 1613 (1955).

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8.3.7 Protactinium-231. This isotope is found in all uranium nimerals where it occurs as a member of the actinouranium decay chain. The history of protactinium and its relationship to the rest of the actinouranium family is described thoroughly in Chapter 6.

Protactinium-231 is an alpha-emitter with a half-life of 34,800 ± 300 years as recalculated by ELSON[1] from the data of VAN WINKLE, LARSON, AND KATZIN.[2] It is the longest-lived of the isotopes of protactinium.

The alpha spectrum of Pa^{231} is complex so that magnetic spectrometers of high resolution are required to make a significant analysis of it. The results of three recent studies are shown in Table 8.22. It can be seen that each succeeding study has resulted in the resolution of more groups. It seems certain that further groups will be revealed by future studies although the low specific activity of Pa^{231} is a deterrent to further work. In Table 8.22, for example, it can be noted that no alpha-group with an abundance less than 1 percent is recorded.

The complexity of the alpha spectrum implies a complex gamma spectrum. Here again, repeated re-examination of the radiations with more care and better instruments has successively revealed more and more transitions. MEITNER[7] first examined the conversion electron spectrum and reported electron energies corresponding to gamma transitions of 95.2, 294, and 323 kev. Her results were later confirmed by HAGGSTROM.[8] SCHARFF-GOLDHABER AND MacKEOWN[9] examined the low-energy photons in proportional counters and found a prominent 27 kev gamma ray in addition to L x-rays with energies of 13, 16, and 20 kev. TEILLAC[10] saw electron tracks in a Wilson cloud chamber corresponding to gamma-rays with energy 44 and 66 kev. RIOU[11] used absorption curves taken with proportional counters and geiger counters to resolve out L x-rays and gamma rays energies of 27, 100, and 300 kev energy. His values for the abundances per 100 alpha disintegrations were 36, 9, 2.5, and 4, respectively, for these photon groups. MOUHASSEB AND RIOU[12] gave slightly different relative abundances. RIOU[11] decided that the 100 kev photon group was a complex mixture of K x-rays and a gamma ray of about 87 kev. The 300 kev gamma ray component was also suspected of being complex. MOORE[13] examined the gamma ray photons with a NaI spectrometer and found peaks at 27, 95, and 280 kev. HUMMEL[6] did a similar study but found a value of 300 Mev instead of 280 for the highest energy peak.

Table 8.22

Alpha Groups of Pa^{231}

ROSENBLUM, COTTON, AND BOUISSIERES[3]		GOL'DIN TRETYAKOV, AND NOVIKOVA[4]		HUMMEL, ASARO, AND PERLMAN[5,6]			
Energy (Mev)	Abundance (%)	Energy (Mev)	Abundance (%)	Energy (Mev)	Abundance (%)	Energy of Ac^{224} level (kev)	Hindrance factor
5.042	11	5.0490	12	5.046	10	0	230
		5.0205	23	5.017	23	29.4	68
5.002	47						
		5.0060	26	5.001	24	46.1	51
		4.9740	1.5	4.971	2.3	76.3	330
4.938	25	4.9420	24	4.938	22	110.1	21
				4.921	2.8	127	130
4.838	3	4.8476	1.5	4.839	1.4	211	73
4.720	11	4.7270	10	4.722	11	329	1.5
		4.7040	0.8	4.696	1.4	356	7.6
4.660	1-3	4.6710	1.3	4.666	2.1	387	3.2

The most detailed information on the gamma transitions has come from recent studies of the conversion electrons using magnetic spectrometers of high resolution. The results of FAIK-VAIRANT[14] are given in Table 8.23. The energies of the transitions are given together with the multipolarity assignments which are based on subshell conversion ratios and K/L conversion ratios. The later results of STEPHENS[15] are also given in Table 8.23.

MOORE[13] carried out alpha-gamma and gamma-gamma coincidence studies and introduced a variable time delay between the two detectors in order to detect possible delayed transitions. In the alpha-gamma experiments he measured a delay half-time of $(3.7 \pm 0.1) \times 10^{-8}$ seconds for the 27 kev photons. TEILLAC, RIOU, AND DESNEIGES[16] and FOUCHER, DICK, PERRIN, AND VARTAPETIAN[17] have also measured the delay in the emission of the 27 kev photons following alpha-emission; their values of the half life are 4.2×10^{-8} and 3.7×10^{-8} seconds, respectively. The multipolarity of the 27 kev transition has been firmly assigned as E1 by many authors on the basis of its conversion coefficient. This assignment, the possibility of some M2 admixture, and possible anomolies in the conversion coefficients and the half-life are discussed by ASARO, STEPHENS, HOLLANDER, AND PERLMAN.[18]

STROMINGER[19] set an upper limit of 1.5×10^{-9} seconds to the half life of the 300 kev gamma ray (or gamma ray complex) in an alpha-gamma delayed-coincidence experiment.

ALBOUY[20] measured conversion electrons in coincidence with alpha particles. HUMMEL[6] performed a few exploratory alpha-gamma coincidence experiments in which the more intense alpha particle groups were singled out in a magnetic spectrometer and the coincident gamma spectrum was measured in a NaI crystal spectrometer. His results are summarized in Table 8.24.

Decay schemes for Pa^{231} have been proposed from time to time on the basis of the available data. A notable publication in this respect is that of FAIK-VAIRANT AND RIOU[21] in 1953. But subsequent data have forced revisions in these decay schemes. It is still impossible to present a revised decay scheme for the complex decay of this isotope with any confidence that it will not require some revision or extension as more precise data become available. However, the alpha spectrum does provide a rather firm skeleton of levels on which to built. Furthermore the rather precise data on the conversion electrons permits one to place many gamma transitions with

Table 8.23

Gamma Ray Transitions in Pa^{231}

Deduced from Study of Conversion Electrons

Results of FAIK-VAIRANT[14]		Results of STEPHENS[15]	
Energy (kev)	Multipolarity assignment	Energy (kev)	Multipolarity assignment
		18.88	M2
		25.31	M1
		27.28	E1 (M2)
		29.88	M2
33.6			
38.0	E2	38.12	
		44.04 (?)	
		52.60	(M2)
56.9	E2	57.08	E2
63.5	E2	63.51	E2
		74.04	E2
82.3	M1 or M2		
		77.22	M1
		96.68	E2
		100.66	E2
102	E2	102.55	E2
198	E2		
259	M1	259.8	
		283.1	
		299.4	
301	M1	302.0	
331	M1	329.2	
357	M1	356.3	
383	E1 or M2		

Note: MOORE[13] reinterpreted several of the conversion shell assignments of FALK-VAIRANT which had the result of removing the 82.3 kev gamma-ray and inserting gamma-rays of 280, 96.6, 118, and 67.5 kev.

Table 8.24

Alpha-Gamma Coincidence Results of HUMMEL[6]

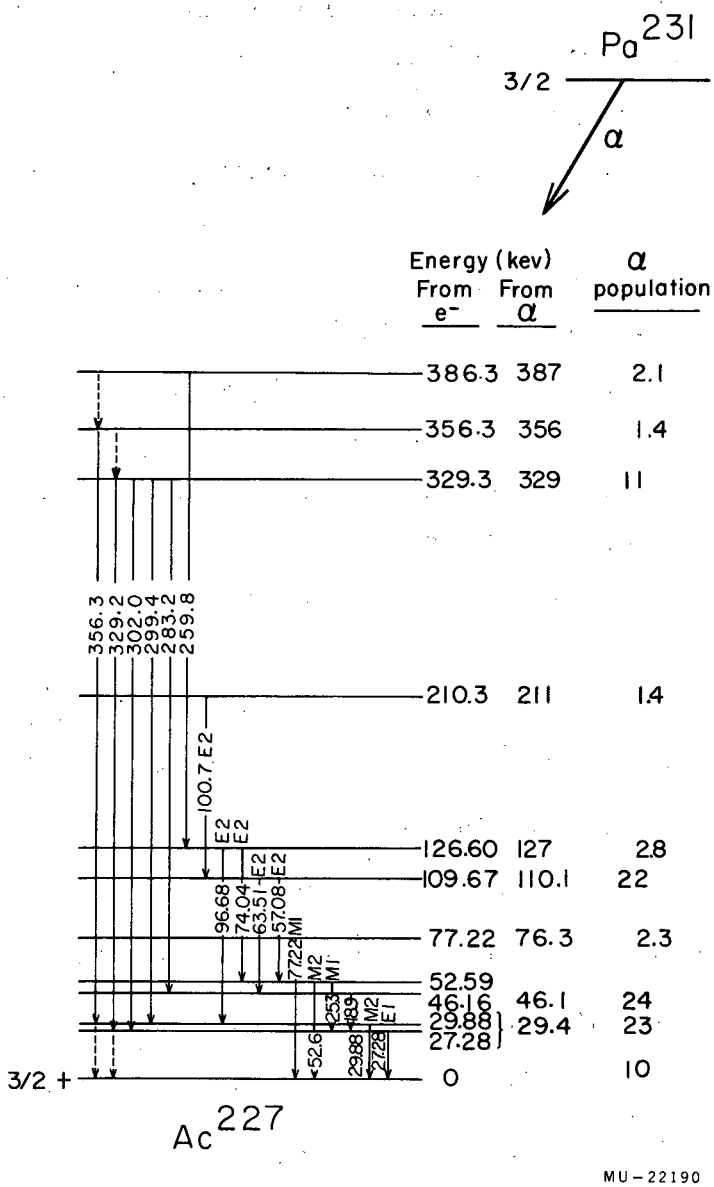
Pa^{231} Alpha-group	Gamma Rays in Coincidence
$\alpha_{29} + \alpha_{46}$	L x-rays + 27 kev γ
α_{110}	L x-rays + 27 kev γ
α_{329}	K x-rays 300 kev γ (complex)
α_{387}	K x-rays ~260 kev γ 300 kev γ ~360 kev γ

considerable confidence. Some of the alternate choices of gamma ray placement can be ruled out by consideration of the available alpha-gamma or gamma-gamma coincidence results. Figure 8.15 shows a tentative decay scheme constructed in this fashion by STEPHENS.[15] One interesting feature is the presence of pairs of close-lying levels near 30 Mev which were not resolved in HUMMEL'S alpha spectrum measurements. It is premature to attempt any interpretation of the decay scheme although some tentative suggestions can be found in a paper by STEPHENS, ASARO, AND PERLMAN.[22]

The nuclear spin of Pa^{231} is 3/2 as determined from atomic spectra.[23] NILSSON AND MOTTELSON[24] discuss the possible NILSSON state assignment of the Pa^{231} ground state and conclude that this state is the 3/2 member of an anomalous K = 1/2 band in which the decoupling constant a in the equation,

$$E = \frac{\hbar^2}{2\mathfrak{I}} \left[I (I + 1) + a (-1)^{I+1/2} (I + 1/2) \delta_{K,1/2} \right],$$

is negative and sufficiently large to drop the 3/2 level below the 1/2 level. (Compare discussion of "Anomalous" K = 1/2 rotational bands in Chapter 3.) The $[N\ n_z\ \Lambda]$ quantum numbers are [530]. This assignment is based on a consideration of the common features of the decay of Pa^{231} and Pa^{233} and to the results of experiments on the excitation of levels in Pa^{231} by the Coulomb excitation process.[25] A similar assignment is made by STEPHENS, ASARO, AND PERLMAN[22] on similar considerations.



8.15 Decay scheme for Pa^{231}.

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8.3.8 Protactinium-232. This nuclide may be made by the following reactions*:

$$Th^{232}\ (d,2n)\ Pa^{232}$$
$$Th^{232}\ (p,n)\ Pa^{232}$$
$$Th^{232}\ (\alpha,p3n)Pa^{232}$$
$$Pa^{231}\ (d,p)\ Pa^{232}$$

An additional method of preparation is the neutron irradiation of Pa^{231},

$$Pa^{231}\ (n,\gamma)\ Pa^{232} \quad \sigma = 293 \text{ millibarns.}$$

This last is the best method for the preparation of samples for detailed study of the Pa^{232} radiations since no Pa^{233} contamination is produced.

GOFMAN AND SEABORG[1] were the first to produce and study this isotope. Protactinium-232 is a beta emitter with a half life of 1.32 days.[2] Several groups of investigators[3-6] have contributed to a preliminary study of the very complex radiations of Pa^{232}. A quite definitive study has been carried out by BJORNHOLM, KNUTSEN, AND NIELSEN[7] and we quote here almost exclusively from their work.

Approximately 60 conversion lines have been identified. These lines are superimposed on a complex beta spectrum. The total intensity of the conversion electrons is greater than the beta spectrum electron intensity. Accurate measurement of the conversion lines was made on a six-gap beta spectrometer with resolution adjusted to 0.2 percent. The absolute accuracy of the energy determinations was 0.2%. The gamma transitions deduced from the conversion lines are listed in Table 8.25. This table also shows the multipolarity assignments made on the basis of $L_I + L_{II}/L_{III}$ and K/L electron ratios, and K-conversion coefficients. The Fermi analysis of the beta spectrum made by these workers is shown in Table 8.26. Valuable information on the arrangement of these gamma rays into a decay scheme was obtained by beta-gamma and e-γ coincidence measurements. In these experiments the gating signal was provided by pulses from the detector of the six-gap beta spectrometer with the spectrometer focused on a specific electron line. The coincident gamma spectrum was measured with a sodium iodide crystal in combination with a multichannel analyzer.

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*An indication of the cross sections is given in Table 5.9 of Chapter 5.

Table 8.25

Gamma Transitions in the Decay of Pa^{232}

Energies, Multipolarities, and Intensities

E_γ kev	Multi-polarity	L_{I-II}/L_{III} exp.	L_{I-II}/L_{III} theor.	K/L exp.	K/L theor.	γ-Intensity (%) from total γ-spectrum	γ-Intensity (%) from coinc. γ-spectra	γ-Intensity (%) calc. from conv. data	γ-Intensity (%) adopted value	total intensity %
47.5	E2	1.2	1.2			< 1		0.2		78
K-Xray						9.6	2.5	3.0	2.5	3.5
105.4	E1									
109.0	E2	1.9	1.7					3.3		26
150.2	E1	3.4	4.1	3.4	5.0	12	13	12	12	14
388.0	M1+	4.0	250	2.1	4.6	4.8	7.3	0.5	0.5^a	8.0
	E2		4.4		1.3			6.7	6.7^a	
422.5	M1+			≈3	4.6	≤ 3	2.5		0.5^a	2.8
	E2				1.5				2.0^a	
454.4	M1+	10	260	2.6	4.6	8.4	5.0	1.1	1.3^a	5.7
	E2		5.8		1.6			6.3	3.7^a	
472.6	E1			≈2	5.2		4.2	4.0	4.1	4.2
516.1	E1					2.4	3.3	≈4	3.3	3.4
564.5	E1			4	5.2	6.0		2.3		2.3
583.8	E1				5.2		6.4	6.0	6.2	6.3

(continued)

Table 8.25 continued

E_γ kev	Multi-polarity	L_{I-II}/L_{III} exp.	L_{I-II}/L_{III} theor.	K/L exp.	K/L theor.	γ-intensity % from total γ-spectrum	γ-intensity % from coinc. γ-spectra	γ-intensity % calc. from conv. data	γ-intensity % adopted value	total intensity %
676.5	E0			3				0	0	0.017
687.5	E0			3.6		< 3		0	0	0.062
692.9	E0			4				0	0	0.032
711.6	E2					< 2	< 0.5	0.23	0.23	0.23
757.0	E2			3	3.1	< 2	< 1	0.67	0.67	0.68
819.6	E2			3.3	3.3			8.2	8.2	8.3
865.3	E2			3.3	3.5	30		2.8	2.8	2.8
868.0	E2							6.3	6.3	6.4
894.8	E1			6.0	5.3		21	21	21	21
971.0	E1			5.8	5.4	40		42	41	41

[a]The adopted values for the three mixed M1 + E2 transitions are calculated by combining K and L conversion line intensities with the gamma-intensity found from coincidence spectra. Intensities are expressed in percent of beta decays. This table prepared by Bjørnholm, Knutsen and Nielsen.[7]

Table 8.26

Fermi analysis of the β-spectrum

Q_β = 1345 ± 20 kev

Max energy kev	Intensity	Leading to excited levels at: kev
1295 ± 20	0.7 %	47.5 ± 20 [a]
1190 ± 20	0.8 %	156.5 ± 20 [a]
600 ± 100	< 1 %	(750 ± 100)
320 ± 30	98 %	1025 ± 30

Table prepared by Bjørnholm, Knutsen and Nielsen.[7]

[a] The value of Q_β is based on the assumption that the two high energy β-groups populate the 2+ and 4+ members of the ground-state rotational band at 47.5 kev and 156.5 kev. This is suggested by the energy difference, 105 ± 20 kev; and by the observation that high energy β-rays are only coincident with low energy (conversion) electrons and weak γ-rays of ≈ 109 kev.

The decay scheme as formulated by BJORNHOLM, KNUTSEN, AND NIELSEN[7] is shown in Figure 8.16. The more intense gamma rays can be placed, without benefit of the coincidence experiment results, by the following considerations. Uranium-232, like all even-even nuclei in this region of mass numbers, has a deformed shape and its lowest levels of excitation represent rotational excitation with a spin sequence 0+, 2+, 4+, etc. This ground state rotational band is known to occur in U^{232} from a study of the alpha-decay of Pu^{236} (See Section 9.2.6 of Chapter 9). Hence in the decay of Pa^{232} there is no question about the placement of the intense E2 transitions with energy 47.5 kev and 109 kev. HILL, HOLLANDER, AND PERLMAN[8] have measured the precise values 47.48 and 108.9 kev for these energies. The next step is to note that many of the more energetic transitions occur in pairs which differ in energy precisely by the 47.5 or 108.9 kev difference of the levels of the ground state rotational band. Each such pair of transitions can be used to set the energy of an excited level; for example the levels at 564, 630, 868, and 913 kev are fixed with certainty in this way. The energies of other transitions are then found to be equal to the energy difference of two of these higher levels and can be placed in the scheme. In the case of the three levels at 693, 735, and 833, which are labeled beta-vibrational band in the figure, the additional information obtained from $e^-\gamma$ and e^-e^- coincidence experiments was crucial in the proof of their correct placement in the scheme. The multipolarity assignments of the gamma rays were used in the assignment of spins and parities to all levels. It is beyond the scope of this summary to include all the experimental results and detailed arguments which entered into the construction and interpretation of the Pa^{232} decay scheme, but these can be found in the original paper.[7] We shall, however, call attention to several interesting features of the level system of U^{232}.

The U^{232} level system is an excellent example of the types of collective excitation described by the collective model of the nucleus. There is first the ground state rotational band with the level spacings, the spins and parity sequence, and the pattern of electric quadrupole cascade de-excitation which are universally observed in the mass region above mass 225. At 564 and 630 kev there are levels with spin and parity 1- and 3-, respectively. These are interpreted as an excitation of an

octupole vibrational mode. The K-quantum number of this excitation is zero. The 1- level is the base state for one phonon of vibrational excitation and the 3- level is the first rotational excitation of this base state. Both states de-excite by a pair of E1 transitions to two members of the ground state rotational band. According to theory the ratio of the reduced transition probabilities should be given by ratios of squares of Clebsch-Gordan coefficients. This proves to be the case as shown in Table 8.27. The levels at 693, 735, and 833 kev are believed to be a rotational band of levels based on a 693 kev level which represents the beta-vibrational mode of quadrupole collective excitation. These levels decay by E2 and E0 transitions to the ground state rotational band. These levels are very weakly populated if at all by direct beta decay. This fact receives a natural explanation in terms of K-forbiddenness since the spin and K-value of Pa^{232} is most likely 3. The weak population of the levels of the beta-vibrational band makes it difficult to identify with certainty all the E2 transitions which de-excite them or to determine reduced transition probability ratios. The gamma-vibrational mode of quadrupole excitation is represented by the levels at 868 and 913 kev. Theory predicts that these levels will de-excite by E2 transitions to members of the ground state band. This de-excitation is observed and the ratios of reduced transition probabilities for pairs of these E2 transitions agree with the theoretical calculation from Clebsch-Gordan coefficient ratios. See Table 8.27. The nature of the very heavily populated levels of 1018 and 1052 kev is not identified with certainty. Between them they receive about 98% of the total beta intensity.

One puzzling feature of the decay scheme is the existence of several transitions from levels in the gamma-vibrational band to levels in the beta-vibrational band.

Table 8.27

Reduced Branching Ratios

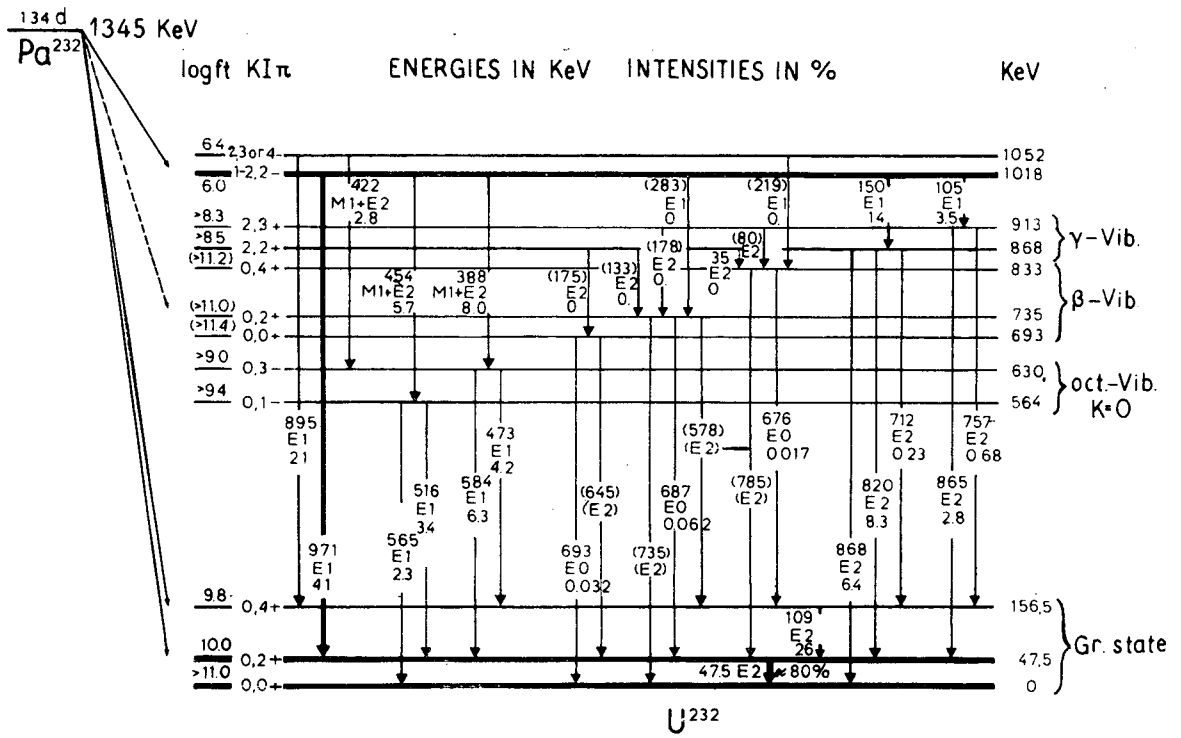
Upper level		Multipolarity	Lower levels		Branching intensities	
kev	K,I,π		kev	K,I,π	exp.	theor.
564.0	0,1-	E1	0	0,0+	1	1
			47.5	0,2+	1.9 ± 0.2	2.0
630.2	0,3-	E1	47.5	0,2+	1	1
			156.5	0,4+	1.24 ± 0.05	1.33
868.0	2,2+	E2	0	0,0+	0.69 ± 0.05	0.70
			47.5	0,2+	1	1
			156.5	0,4+	0.057±0.006	0.05
912.8	2,3+	E2	47.5	0,2+	1	1
			156.5	0,4+	0.47 ± 0.10	0.40
1018.3	2,2-	E1	868.0	2,2+	1	1
			912.8	2,3+	0.6 ± 0.1	0.50
same	1,2-	M1	564.0	0,1-	1	1
			630.2	0,3-	0.6 ± 0.1	0.67
same	1,2-	E2	same	0,1-	1	1
				0,3-	4 ± 1	4.0

Table 8.28

Rotational Bands

$$E_{rot} = A\,(I\,(I+1)) + B\,(I\,(I+1))^2$$

Band head		$\frac{\hbar^2}{2\,\Im} = A$	$-B$
kev	K,I,π	kev	kev
0	0,0+	7.86 ± 0.03	0.007 ± 0.002
564	0,1-	6.6 ± 0.1	
693	0,0+	7.0 ± 0.2	0.002 ± 0.006
868	2,2+	7.5 ± 0.1	



8.16 The decay scheme of Pa^{232} as formulated by Bjornholm, Knutsen, and Nielsen. The thickness of the lines reflects in a rough way the intensities of the transitions or the extent to which each level is populated. The log ft values were determined from the in-out intensity balance of each level from which the intensities of the individual beta branches were calculated. The β- branches to the ground state band were, however, measured directly. The gamma energies given in parentheses are very weak in intensity and are not identified with certainty.

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3. C. I. Browne, Jr., D. C. Hoffman, H. L. Smith, M. E. Bunker, J. P. Mize, J. W. Starner, R. L. Moore, and J. P. Balagna, Phys. Rev. 96, 827A (1954).

4. Ong Ping Hok, Thesis, Free University of Amsterdam, March, 1955.

5. Ong Ping Hok and G. J. Sizoo, Physica 20, 77 (1954).

6. M. W. Hill, Thesis, University of California, 1958; also published as University of California Radiation Laboratory Report UCRL-8423, August 1958.

7. S. Bjørnholm, A. Knutsen, and O. B. Nielsen, to be published, 1961.

8. M. W. Hill, J. M. Hollander, and I. Perlman, unpublished results (1956).

8.3.9 Protactinium-233. The production of Pa^{233} by neutron irradiation of thorium and its original identification as a 27.4 day beta emitter are mentioned in the discussion of the 4n + 1 series of radioactive elements in Section 7.1 of the preceding chapter. As a precursor of the important 4n + 1 series isotope, U^{233}, it is an important collateral member of the 4n + 1 series.

The radiations of Pa^{233} are complex. There are numerous gamma rays ranging in energy from 28 to 416 kev. Many of these give rise to strong conversion electron lines and Auger electrons superimposed on a complex beta spectrum. Gamma ray spectrum analysis by the sodium iodide scintillation spectrometer technique is not very useful because of the complexity of the spectrum and the limited resolving power of the method. Photon energies can be accurately measured in bent crystal spectrometers. The L x-ray and gamma ray photons up to 100 kev were measured by BROWNE AND PERLMAN[1] with such a spectrometer; their values for four gamma rays are listed in Table 8.29. Several years later this study was repeated and extended to much higher energies by GALLAGHER, ALBRIDGE, AND HOLLANDER;[2,11] their results are also listed in the table. Many authors have contributed to the accurate measurement and interpretation of the conversion electron spectrum of Pa^{233} and except for a few doubtful points the agreement on the major transitions has been gratifying. The gamma ray transition energies reported by several authors are summarized in Table 8.29. The multipolarity assignments of BISGARD, DAHL, AND OLESEN,[16] of ALBOUY AND VALADARES[17] and of ALBRIDGE AND HOLLANDER[11] are summarized in Table 8.30.

It has been difficult to make a detailed analysis of the beta spectrum because so many conversion electrons are superimposed on the beta continuum; attempts at such a resolution were made by BRODIE[5] and by ONG PING HOK[3,4] with results which agree very well as to end-point energies but only roughly as to relative intensity. BRODIE'S[5] values are: 568 ± 5 kev (5 percent), 256 ± 4 kev (57 percent) and 140 ± 14 kev (38 percent). Each of these groups may be unresolved mixtures of two groups separated by 16 to 40 kilovolts. The beta component intensities and log ft values shown in the decay scheme were derived from gamma ray transition intensities, which clearly seem to indicate this greater complexity in the beta spectrum.

The gamma transitions listed in Tables 8.29 and 8.30 can be arranged in the decay scheme of Figure 8.17. This scheme was prepared by ALBRIDGE AND

Table 8.29 Energy (in kev) of gamma transitions of Pa^{233}

	From measurements of conversion electrons					Crystal spectrometer photon measurements	
	Ong Ping Hok[3,4]	Brodie[5]	Elliott and Underhill[6]	Keller and Cork[7]	Albridge, Hill, and Hollander[8,11]	Gallagher, Albridge, and Hollander[2]	Browne and Perlman[1]
1	15.8	--	--	--	17.26	--	--
2	27.5	28.6	--	28.9	28.54	--	28.7(100)
3	40	40.7	--	40.6	40.29	40.35	40.5(75)
4	--	--	--	--	--	41.65	--
5	58	58	--	58.1	57.90	--	--
6	75	75.6	76	75.7	75.13	75.28	75.4(3)
7	86	87	88	87.1	86.45	86.59	87.0(3)
8	104	104	105	104.5	103.6	103.86	--
9	--	--	--	--	--	145.42	--
10	272	--	--	272.6	271.5	271.62	--
11	301	302	298	301.5	299.8	300.20	--
12	313	314	310	313.1	311.7	311.91	--
13	341	343	339	342	340.3	340.51	--
14	376	--	--	376.5	375.5	375.35	--
15	400	--	398	399.9	398.3	398.57	--
16	417	--	415	416.4	415.6	415.87	--
17	476	--	474	--	--	--	--

Table 8.30 The decay of Pa^{233} -- multipolarity assignments of various workers

E_γ (kev)	Conversion ratio	Experimental values Bisgard et al.[b]	This work	Theoretical values[a]			Multipolarity		
				E1	E2	M1	Bisgard et al.[a]	Albouy and Valadares[c]	Albridge and Hollander[d]
28	$\frac{M_I + M_{II}}{M_{III}}$	~9	4.3	2.2	0.7	170	M1 + E2	M1 + E2	98% M1 2% E2
	$\frac{M_{III}}{M_{IV}}$	>20	--	1.9	62	7.6			
40	$\frac{L_I + L_{II}}{L_{III}}$	1.0	1.7	1.1	1.1	230	M1 + E2	20% M1 80% E2	70% M1 30% E2
	$\frac{L_I}{L_{II}}$	~0.4	0.27	0.7	0.03	13			
	$\frac{M_I + M_{II}}{M_{III}}$	1.7	--	2.0	1.1	200			
	$\frac{M_{III}}{M_{IV}}$	$\gtrsim 50$	--	2.8	46	7.3			
76	$\frac{L_I + L_{II}}{L_{III}}$	>130	74	2.2	1.3	223	M1	M1($\ll$1% E2)	98% M1 2% E2
	$\frac{L_I}{L_{II}}$	~13	7.1	1.4	0.06	12			

(continued)

Table 8.30 - continued

E_γ (kev)	Conversion ratio	Experimental values Bisgard et al.[b]	This work	Theoretical values[a]			Multipolarity		
				E1	E2	M1	Bisgard et al.[a]	Albouy and Valadares[c]	Albridge and Hollander[d]
87	$\frac{L_I + L_{II}}{L_{III}}$	>20	40	2.4	1.3	207	M1	M1(< 1% E2)	97% M1 3% E2
	$\frac{L_I}{L_{II}}$	~9	7.4	1.5	0.06	11			
104	$\frac{L_I + L_{II}}{L_{III}}$	>30	30	1.9	1.6	229	M1	M1 (< 1% E2)	95% M1 5% E2
	$\frac{L_I}{L_{II}}$	~9	5.6	0.9	0.06	11			
273	K/L	0.55	$\geq$ 0.59	5.2	0.67	4.6	E2	E3,E4, or E5 or M1 + E2	E2
	$\frac{L_I + L_{II}}{L_{III}}$	2.8	$\geq$ 1.3	6.7	3.3	274			
301	K/L	5.0	4.1	5.2	0.84	4.5	M1	M1	90% M1 10% E2
313	K/L	5.1	3.9	5.1	0.9	4.5	M1	M1	M1
341	K/L	5.0	4.0	5.2	1.1	4.5	M1	M1	95% M1 5% E2
	$\frac{L_I + L_{II}}{L_{III}}$	>100	--	8.2	4.2	277			

continued

Table 8.30 - continued

E_γ (kev)	Conversion ratio	Experimental values Bisgard et al.[b]	This work	Theoretical values[a]			Multipolarity		
				E1	E2	M1	Bisgard et al.[a]	Albouy and Valadares[c]	Albridge and Hollander[d]
377	K/L	1.2	0.90	5.1	1.2	4.6	E2	--	E2
	$\frac{L_I + L_{II}}{L_{III}}$	3.1	3.0	9.0	4.9	287			
400	K/L	~2.4	1.0	5.2	1.3	4.5	E2	--	E2
	$\frac{L_I + L_{II}}{L_{III}}$	4.0	5.1	9.2	5.1	294			
417	K/L	3.9	2.8	5.1	1.5	4.4	M1	--	22% M1 78% E2
	$\frac{L_I + L_{II}}{L_{III}}$	>20	9.0	9.9	5.5	297			

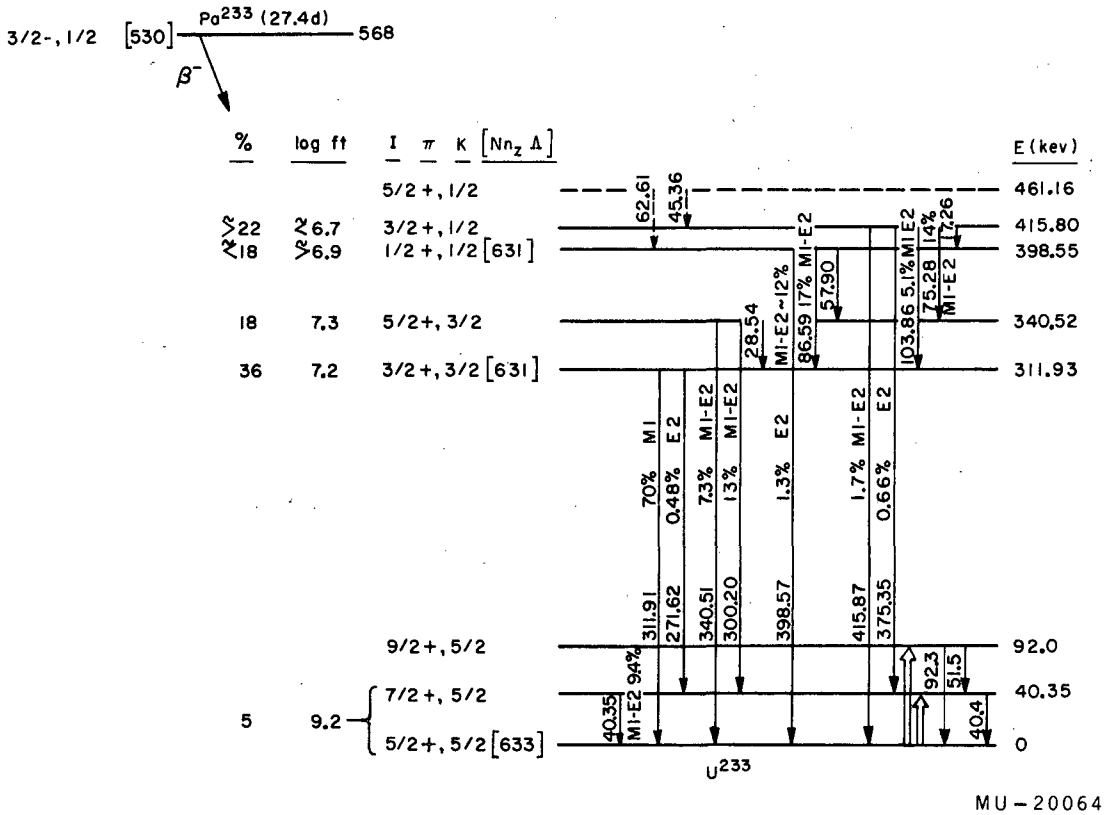
This table is a copy of one presented by Bisgard et al., with the data of other workers included for comparison.

a. M. E. Rose, Internal Conversion Coefficients 1958.

b. Bisgard, Dahl, and Olesen, Nuclear Physics 12, 612 (1959).

c. Albouy and Valadares, J. Phys. Radium

d. Albridge and Hollander, UCRL-8642.



8.17 A tentative decay scheme for Pa^{233}. The percent beta populations listed were deduced by Albridge et al.[13] from the gamma transition intensities which were in turn deduced from relative electron and photon intensities. The log ft values were calculated from the beta populations by the method of Moszkowski. The level at 461.16-kev was assigned on the basis of two weak electron lines which could be assigned to transitions of 62.11- and 45.36-kev. Albridge et al.[13] also discuss the possible assignments of two weak photon transitions of 41.65- and 145.42-kev to a level at 270.3- or 457.4-kev.

HOLLANDER[11] but in general features it is based on the earlier scheme of BRODIE[5] which was supported by HOK AND KRAMER.[4] This scheme accounts for all the gamma rays listed except for the doubtful weak ray of 476 kev energy and gives a good "in-out" intensity balance for each level. The scheme receives support from additional pieces of evidence. ELLIOTT AND UNDERHILL[6] observed coincidences between the conversion electrons of the 312 kev transition and those of the 75, 87, and 104 kev transitions. UNIK[12] performed similar electron-electron coincidence experiments with similar results. He also showed that the 75 kev transition is in coincidence with a 151 ± 4 kev beta group and the 86 kev transition is in coincidence with a 166 ± 5 kev beta group. The beta spectrum in coincidence with the 312 kev transition is complex and comprises beta groups with end point energies of 250 ± 5 kev and 168 ± 8 kev.

The nuclear spin of Pa^{233} has been determined to be 3/2 by the atomic beam resonance technique by WINOCUR.[15] The Nilsson orbital assignment for the ground state is believed[13] to be 1/2- [530] where these numbers represent the following quantum numbers, $K\ \pi\ [N\ n_z\ \Lambda]$. The rotational band based on this K = 1/2 level has an anomalous ordering and the I = 3/2 member of the band lies lowest. The arguments upon which this assignment is based are cited in the discussion of the decay scheme of Np^{237} in Chapter 9, Section 9.1.9.

The Nilsson assignments shown in the diagram for the levels of U^{233} follow those given by NEWTON.[9] He contributed greatly to the identification of the lowest-lying levels in U^{233} by Coulomb excitation of levels at 40.1 and 92 kev and by identification of these as the 7/2 and 9/2 rotational states of a K = 5/2 ground state. This Coulombic excitation is shown in the figure by upward-pointing arrows. The ground state of U^{233} is known to have spin 5/2 from atomic spectra and paramagnetic resonance studies. (See Section 8.4.6.) NEWTON[9] assigned the Nilsson wave function 5/2+ [633] to the ground state and this is considered to be well established. NEWTON'S identification of the levels at 312 and 398 kev with Nilsson wave functions were made with much less certainty at the time but later data on the multipolarity assignments[11,16] of the gamma rays in the decay of Pa^{233} have served to confirm them. See especially the discussion of ALBRIDGE.[11] Other discussions of the U^{233} Nilsson assignments can be cited.[13,14]

If these Nilsson assignments are accepted as correct, both for Pa^{233} and U^{233}, it is possible to interpret many details of the decay scheme in terms of the selection rules pertinent to the collective model of the nucleus. For example the high log ft values for beta transitions to the ground state rotational band are explained by K-forbiddenness and violation of the selection rules in the asymptotic quantum numbers. Also many of the branching ratios in the gamma ray de-excitation of a higher level of U^{233} can be calculated theoretically by Clebsch-Gordan coefficient ratios. Details of this type are discussed by ALBRIDGE.[11]

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1. C. I. Browne and I. Perlman, unpublished results quoted in Rev. Mod. Phys. 25, 469 (1953).
2. C. J. Gallagher, R. G. Albridge, and J. M. Hollander, Phys. Rev. to be published.
3. Ong Ping Hok, Thesis, University of Amsterdam, March 1955.
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5. W. D. Brodie, Proc. Phys. Soc. 67A, 397 (1954).
6. L. G. Elliott and A. B. Underhill, Atomic Energy Research Establishment Report, (Harwell) HAR-761 (1952).
7. H. B. Keller and J. M. Cork, Phys. Rev. 79, 1030 (1950).
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10. M. W. Hill, Thesis, University of California 1958; also published as University of California Radiation Laboratory Report, UCRL-8423, August 1958.
11. R. G. Albridge and J. M. Hollander, unpublished results; R. G. Albridge, Thesis, University of California 1960; also published as University of California Radiation Laboratory Report, UCRL-8642, 1960.
12. J. Unik, Thesis, University of California 1960; also published as University of California Radiation Laboratory Report, UCRL-9105, 1960.

13. F. S. Stephens, F. Asaro, and I. Perlman, Phys. Rev. 113, 212 (1959).

14. B. R. Mottelson and S. G. Nilsson, Mat. Fys. Skr. Dan. Vid. Selsk 1, 8 (1959).

15. J. Winocur, Thesis, University of California, 1960; also published as University of California Radiation Laboratory Report, UCRL-9174, April 1960.

16. K. M. Bisgard, P. Dahl, and K. Olesen, Nuclear Physics 12, 612 (1959).

17. G. Albouy and M. Valadares, J. Phys. radium, to be published (1960).

8.3.10 Protactinium-234. In the decay chain of U^{238}, the species Pa^{234} occurs in two isomeric forms known as UX_2 and UZ. UX_2 has a half-life of 1.18 minutes and UZ has a half-life of 6.7 hours; both are beta-emitters. The discovery of these interesting nuclides during the early history of radioactivity is mentioned in Section 6.1 of Chapter 6, and the genetic relationships within the uranium decay chain are covered in Section 6.2 of that chapter. The radiations of UX_2 and of UZ are usually studied in a mixture of the UX_1-UX_2-UZ complex and it is convenient to discuss the radiations of all three nuclides in one place; hence the radiations of the isomers of Pa^{234} are discussed in Section 8.2.12 entitled Thorium-234 and the UX_1-UX_2-UZ Complex.

8.3.11 Protactinium-235. HARVEY AND PARSONS[1] prepared Th^{235} and Pa^{235} by neutron irradiation of Th^{234}(UX_1). The reactions are:

$$Th^{234}\ (n,\gamma)\ Th^{235} \xrightarrow{\beta^-} Pa^{235}.$$

The 24-day Th^{234} was extracted from one kilogram of natural uranium and thoroughly separated from uranium and protactinium immediately before neutron irradiation. Ten minutes after the completion of the irradiation, a protactinium fraction was purified and examined. A 23-minute beta-emitting protactinium isotope which could only be Pa^{235} was found. From milking experiments, an upper limit of 5 minutes was set on the half-life of Th^{235}.

MEINKE AND SEABORG[2] prepared Pa^{235} by bombardment of uranium with 19-Mev deuterons and 9.5-Mev protons:

$$U^{238}\ (d,\alpha n)\ Pa^{235} \qquad \sigma \approx 2 \times 10^{-27}\ cm^2$$

$$U^{238}\ (p,\alpha)\ Pa^{235} \qquad \sigma \approx 3 \times 10^{-29}\ cm^2$$

They reported a half-life of 23.7 minutes. The beta end-point energy was set at 1.4 Mev by absorption measurements. No gamma rays were observed. LINDNER AND OSBORNE[3] have prepared Pa^{235} by proton bombardment of uranium at higher energies. They report the following cross sections: 100 Mev, 5.7 mb; 175 Mev, 7.3 mb; 250 Mev, 15 mb; 340 Mev, 21 mb. It remains to be determined whether the decay of Pa^{235} involves the 26 minute isomer of U^{235} (See Section 8.4.8).

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2. W. W. Meinke and G. T. Seaborg, Phys. Rev. 78, 475 (1950).

3. M. Lindner and R. N. Osborne, Phys. Rev. 103, 378 (1956).

8.3.12 Protactinium-237. CRANE AND IDDINGS[1] bombarded uranium with high energy deuterons (40 - 190 Mev) and found in the protactinium fraction the 23.5-minute Pa^{235} produced by the (d,αn) reaction and in addition, a small amount of a 10.5 ± 1 minute beta emitter which they assigned to Pa^{237}, formed by the reaction:

$$U^{238}\ (d,2pn)\ Pa^{237}$$

The isotope identification and the half life measurement were made by the repeated separation of U^{237} daughter activity.

TAKAHASHI AND MORINAGA[2] prepared Pa^{237} by the bombardment of uranium with a 25 Mev bremsstrahlung beam

$$U^{238}\ (\gamma,p)\ Pa^{237}$$

In the decay of a radiochemically purified protactinium fraction they observed a prominent component with a half life of 39 ± 3 minutes. It is difficult to understand this discrepancy in half life values, but the determination of TAKAHASHI AND MORINAGA is the more direct.

These authors studied the beta spectrum with an anthracene scintillation spectrometer with the results summarized in Table 8.31. They studied the complex gamma spectrum with a sodium iodide scintillation spectrometer and found the gamma rays listed in Table 8.32. They performed a few gamma-gamma coincidence experiments. The low source strengths made it impossible to study the conversion electron spectrum. TAKAHASHI AND MORINAGA constructed the decay scheme shown in the Figure 8.18. The Nilsson orbital assignments were based almost entirely on an examination of Nilsson diagrams and by examination of systematic trends in Nilsson orbitals in neighboring nuclei. Hence it is a highly tentative decay scheme, useful as a guide to future work.

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1. W. W. T. Crane and G. M. Iddings, Phys. Rev. 95, 1702 (1954).

2. K. Takahashi and H. Morinaga, Nuclear Physics 15, 664 (1960).

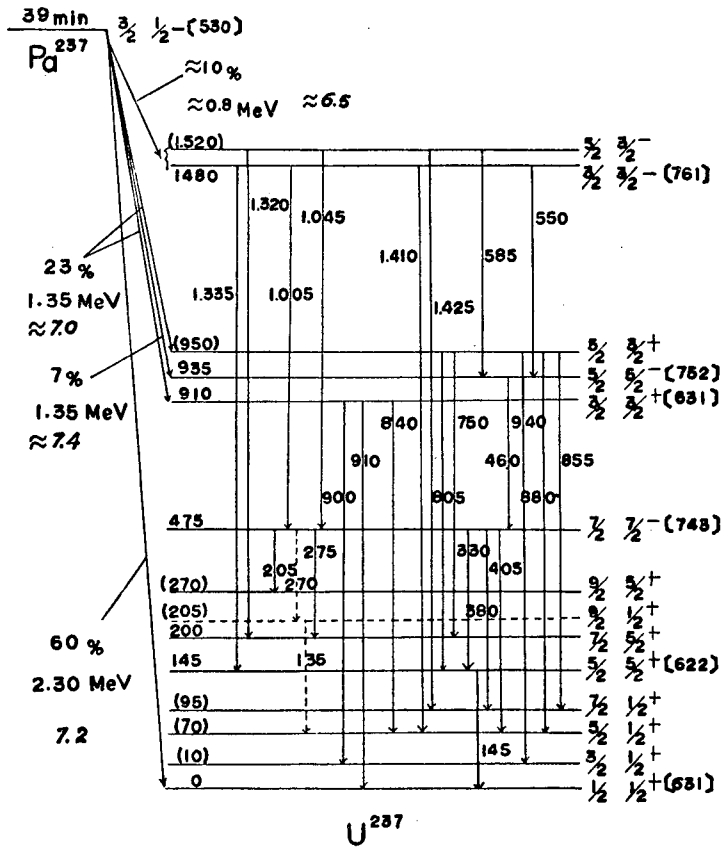
Table 8.31 Beta Groups of Pa^{237}

E_o (Mev)	Intensity (%)	Log ft	Classification
3.30	60	7.2	1 u
1.35	23	7.3	1 u
	7	7.4	a h
≈ 0.8	$\lesssim 10$	$\gtrsim 6.5$	a h

Table 8.32 Gamma Rays of Pa^{237}

Energy (kev)	Relative intensity	Relative intensity coincident with 145 kev gamma-ray
90	50	5
145	45	3
205	55	3
275	20	2.5
330	40	1.5
405	30	(5)*
460	100	3.5
550	30	--
590	25	(5)*
750	50	2.5
805	45	
860	100	
880		
915	100	
1045	35	
1320	10	
1420	15	

*Possibly caused by Pa^{234}(UZ) contamination.



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8.18 Decay scheme of Pa^{237} as formulated by Takahashi and Morinaga.

8.4 THE ISOTOPES OF URANIUM

8.4.1 Uranium-227. This isotope is the parent activity of the U^{227} collateral series which is discussed in Section 7.2.8 of the previous chapter. Our knowledge of U^{227} is limited to the facts that it emits alpha-particles of 6.8 ± 0.1 Mev energy and it has an apparent half-life of 1.3 ± 0.3 minutes.

1. W. W. Meinke, A. Ghiorso, and G. T. Seaborg, Phys. Rev. 85, 429 (1952).

8.4.2 Uranium-228. This isotope is the parent activity for the U^{228} collateral series discussed in Section 7.2.7 of the previous chapter. Our knowledge of U^{228} is sketchy. It decays ~80 percent by the emission of alpha particles and ~20 percent by capture of an orbital electron. The apparent half-life is 9.3 ± 0.5 minutes. RUIZ reports two alpha particle groups: 6.68 Mev (72%) and 658_8 Mev (28%).

1. W. W. Meinke, A. Ghiorso, and G. T. Seaborg, Phys. Rev. 81, 782 (1951).
2. C. P. Ruiz, F. Asaro, and I. Perlman, unpublished results 1960.

8.4.3 Uranium-229. The 58-minute isotope, U^{229}, is the first member of the U^{229} collateral series. The preparation of U^{229} and the characteristics of all members of the decay series to which it gives rise are treated in Section 7.2.6. Uranium-229 decays by the emission of alpha particles in about 20 percent of its disintegrations. It decays by orbital electron capture in the remaining 80 percent of its decay events. RUIZ reports the energy of the main alpha group as 6.35_5 Mev. The percentage abundance of this group is 65. A second group with 25 kev less energy appears in an abundance of 23%. A 9 percent abundant group is observed with 63 kev less energy.

1. W. W. Meinke, A. Ghiorso, and G. T. Seaborg, Phys. Rev. 81, 782 (1951).

8.4.4 Uranium-230. The 20.8 day alpha-emitter U^{230} is a member of the Pa^{230} collateral series[1] which is discussed in Section 7.2.1. The methods of preparation of samples of U^{230} are treated there. Uranium-230 is always associated with its four short-lived alpha-emitting daughters so that its alpha spectrum is complex as shown in Figures 7.10 and 7.11 presented in Section 7.2.1. Here we are concerned only with the details of the radiations of U^{230} itself.

A study of the alpha spectrum in a magnetic spectrometer of high resolution by ASARO AND PERLMAN[2] revealed the complex structures shown in Table 8.33.

The four gamma rays shown in the decay scheme of Figure 8.19 have been identified and subjected to study with NaI crystal spectrometers and high resolution permanent magnet electron spectrographs.[2-4] An interesting feature of the decay scheme is that the alpha particle group of 5.658 Mev energy seen by ASARO AND PERLMAN[2] must actually be an unresolved pair of alpha particles differing by only 4 kilovolts and populating a 4+ level and 1- level. This decay scheme is supported by many pieces of evidence.

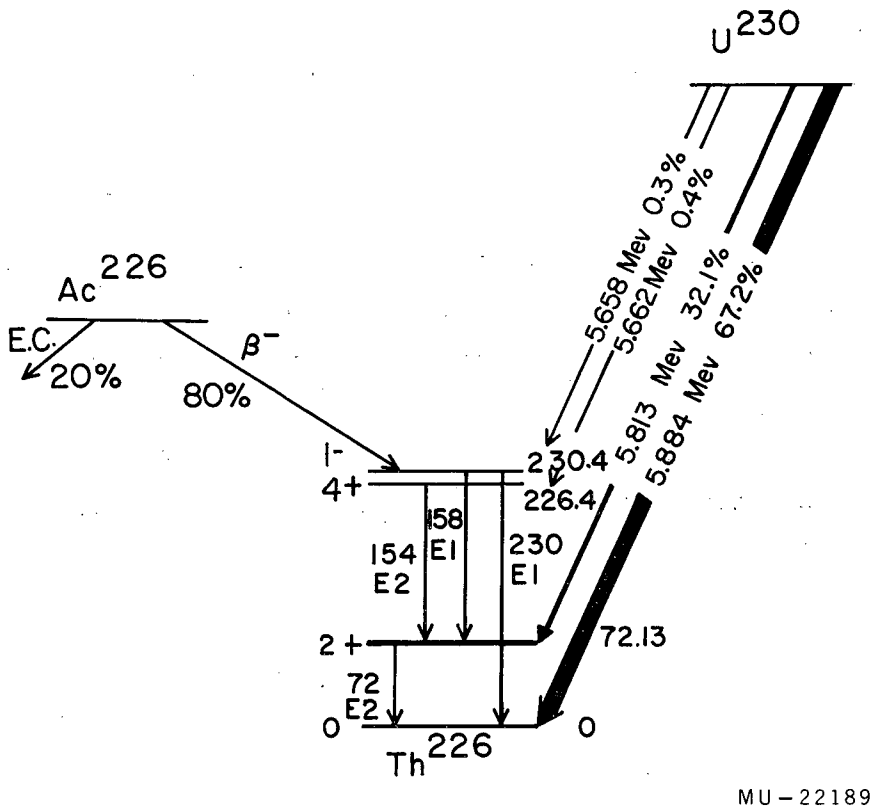
The 0+, 2+, 4+ sequence of levels is the familiar rotational band of levels seen in every even-even nucleus in the heavy element region. The E2 nature of the 72.13 kev and 154.3 kev gamma transitions is shown[4] by the prominent conversion in the L_{II} and L_{III} and in the M_{II} and M_{III} shells and the extremely weak conversion in the L_I and M_I shells. The conversion coefficient of the 72 kev transition as determined by the population of the alpha decay to the state and the number of photons is correct for an electric quadrupole transition.[2] (Alpha population 32.6 percent, 72 kev photons 0.75 percent, $\alpha_K = 42$.)

Angular correlations of alpha particles of a certain energy range with gamma rays of a specific energy were studied by STEPHENS, ASARO, AND PERLMAN.[3] A thin crystal of NaI was used as the alpha detector and a thick crystal was used as the gamma detector. The signals from both detectors were submitted to pulse height analysis for energy discrimination. The observed correlation for α_{72} of U^{230} with the 72 kev gamma ray photons was the correct one for a sequence $0+ \xrightarrow{\alpha} 2+ \xrightarrow{\gamma} 0+$. On the other hand the observed correlation between α_{230} of U^{230} and the 230 kev gamma ray photons was entirely different but in agreement with that to be expected of the sequence $0+ \xrightarrow{\alpha} 1- \xrightarrow{\gamma} 0+$. The 154.1 and 158 kev photons could not be resolved by the NaI crystal

Table 8.33

Alpha Groups of Uranium-230

Group energy (Mev)	Excited state energy (Mev)	Relative abundance (%)
5.884	0	67.2
5.813	72.13	32.1
5.658*	230	0.7
*This is an unresolved doublet consisting of the following two groups.		
5.662	226.4	0.4
5.658	230.4	0.3



8.19 Decay scheme of U^{230} including the beta decay scheme of Ac^{226}.

spectrometer; when the α_{230}- 160 kev gamma angular correlation was studied no variation of counting rate with angle was observed. This is consistent with the supposition that the 230 kev level is a doublet of a 4+ and 1- state and therefore the sequence measured is a mixture of $0+ \xrightarrow{\alpha} 1- \xrightarrow{\gamma} 2+$ and $0+ \xrightarrow{\alpha} 4+ \xrightarrow{\gamma} 2+$.

Other strong evidence for the presence of a 4+, 1- doublet comes from a study[5] of the beta decay of Ac^{226}. This isotope decays to the 1- state in Th^{226} and not at all to the close-lying 4+ state. The relative abundance of the 158 kev gamma ray to that of the 230 kev gamma ray in the decay of Ac^{226} is less by a factor of two than it is in the alpha-decay of U^{230}.

The 1- state in Th^{226} is a result of a collective excitation of the nucleus. This same 1- state appears in the excited levels of many even-even nuclei among the heavy elements and may represent octupole vibrations. See the discussion of Chapter 3.

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1. M. H. Studier and E. K. Hyde, Phys. Rev. 74, 591 (1948).

2. F. Asaro and I. Perlman, Phys. Rev. 104, 91 (1956).

3. F. Stephens, Jr., F. Asaro, and I. Perlman, Phys. Rev. 96, 1568 (1954).

4. W. G. Smith, F. Asaro, and J. M. Hollander, Phys. Rev. 104, 99 (1956).

5. J. Grover, G. T. Seaborg, and F. Stephens, Jr., unpublished information (1954); quoted by Stephens, Asaro, and Perlman, Phys. Rev. 100, 1543 (1955).

8.4.5 Uranium-231. OSBORNE, THOMPSON, AND VAN WINKLE[1] prepared U^{231} by deuteron and helium ion bombardment of protactinium by the reactions:

$$Pa^{231}\ (d,2n)\ U^{231}$$

$$Pa^{231}\ (\alpha,p3n)\ U^{231}$$

It has also been prepared by helium-ion bombardment of thorium by the reaction:

$$Th^{232}\ (\alpha,5n)\ U^{231}$$

In either case it is contaminated somewhat by other uranium isotopes. Uranium-230 is most similar in half-life and hence most likely to interfere in the measurement of the radiations. Uranium-231 decays almost entirely by orbital electron capture with a half-life of 4.2 days. Some preliminary measurements indicate the presence of gamma rays of 51-, 64-, and 76-kev energy. CRANE AND PERLMAN[2] found alpha particles of 5.45-Mev energy in an abundance corresponding to an alpha branching of 5.5×10^{-3} percent. HOLLANDER, STEPHENS, ASARO, AND PERLMAN[3] studied the gamma ray photons and conversion electrons emitted by U^{231} and constructed a decay scheme. This work is discussed together with the studies of the same authors on the beta decay of Th^{231} since in both cases the daughter product is Pa^{231}. See Section 8.2.10.

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1. D. W. Osborne, R. C. Thompson, and Q. Van Winkle, Paper No. 19.11, "The Transuranium Elements," National Nuclear Energy Series, Division IV, Volume 14B, McGraw-Hill Book Company, Inc., New York, 1949.

2. W. W. T. Crane and I. Perlman, unpublished results cited in Rev. Modern Phys. 25, 469 (1953).

3. J. M. Hollander, F. S. Stephens, F. Asaro, and I. Perlman, "Energy Levels of Pa^{231}" paper in publication 1960.

8.4.6 Uranium-232. The uranium isotope U^{232} is a beta-stable, alpha-particle emitter with a half-life of 73.6 years.[1] It was first identified[2] following its growth from the shorter-lived β^- emitter Pa^{232} which had been prepared by the deuteron bombardment of thorium as follows:

$$Th^{232}\ (d,2n)\ Pa^{232} \xrightarrow[\beta^-]{1.32\ \text{days}} U^{232}$$

It has also been produced by other cyclotron-induced reactions:

$$Th^{232}\ (p,n)\ Pa^{232} \xrightarrow{\beta^-} U^{232}$$

$$Th^{232}\ (\alpha,4n)\ U^{232}$$

$$Pa^{231}\ (\alpha,p2n)\ U^{232}.$$

The cross-sections for several of these reactions are given in Table 5.5 of Chapter 5. Uranium-232 can also be prepared by intense neutron irradiation of ionium as follows:

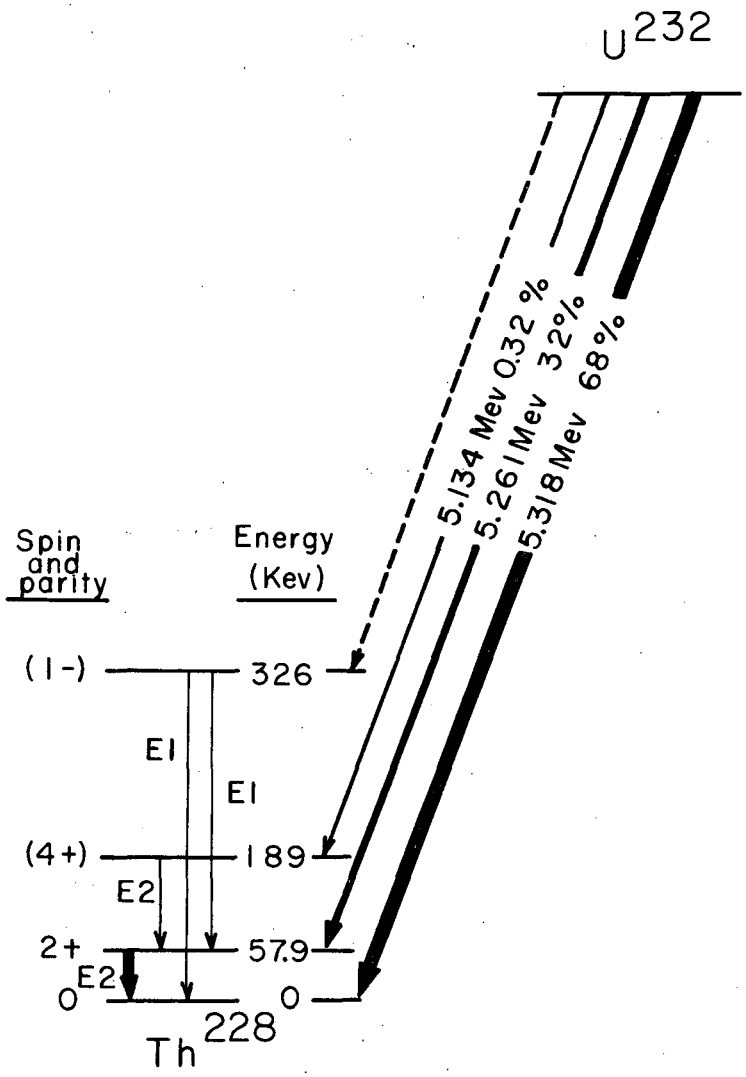
$$Th^{230}\ (n,\gamma)\ Th^{231} \xrightarrow{\beta^-} Pa^{231}\ (n,\gamma)\ Pa^{232} \xrightarrow{\beta^-} U^{232}.$$

Any Th^{232} present in the ionium will cause the formation of considerable U^{233} by a first order reaction but the U^{232} alpha activity may still be predominant because of the longer (1.6×10^5 year) half-life of U^{233}.

A careful study of the alpha spectrum of U^{232} has been carried out by ASARO AND PERLMAN.[3] The gamma radiations have been most thoroughly studied and interpreted by these workers and by SCHARFF-GOLDHABER AND CO-WORKERS.[4]

The alpha spectrum[3] consists of three directly observed groups: 5.318 Mev (68 percent), 5.261 Mev (32 percent) and 5.132 Mev (0.32 percent). A fourth group with energy 4.998 Mev and intensity 0.01 percent is deduced from gamma ray measurements.[3,4] Gamma rays of 57.9, 131, 268, and 326 kev have been observed with the aid of sodium iodide scintillation spectrometers. The photon abundances of these gamma transitions are 0.21 percent, 0.75 percent, 4×10^{-3} percent, and 4×10^{-3} percent, respectively.[3] It will be useful to refer to Figure 8.20 in discussing these alpha and gamma radiations further.

The alpha spectrum of U^{232} follows the familiar pattern for even-even nuclei in this mass region. The principal decay is to the 0+ ground state of Th^{228} with lower abundance groups proceeding to a rotational band of levels based on this ground state. The 131-kev transition is an electric quadrupole transition connecting the 4+ and 2+ levels while the 57.9 kev transition is an electric quadrupole transition from the 2+ level to ground. The electric



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8.20 The decay scheme of U^{232}.

quadrupole nature of both transitions was established by calculating the conversion coefficient from the observed photon intensities and the known alpha group intensities. The cascade arrangement of the 57.9 and 131 kev gamma rays was verified by gamma-gamma coincidence measurements.[3,4]

The level at 326 kev is deduced from the observation of low intensity gamma rays of energy 326 kev and 268 kev which differ by 58 kev. The intensity of these gamma transitions is ~4 x 10^{-3} of the total alpha disintegrations. The 1- spin and parity assignment of the 326 kev level is based on analogy to well characterized 1- levels found in the decay of other even-even alpha emitters in this region such as U^{230}, Th^{226}, and Th^{228}.

The beta decay of $MsTh_2$(Ac^{228}) also gives information on the levels of Th^{228}. The studies of KYLES, CAMPBELL, AND HENDERSON,[5] and of BRODIE[6] indicate that E2 transitions of 56.75 kev and 127.5 kev also appear in the decay of Ac^{228}. A puzzling feature of the 58-kev transition is that two groups[6,8] of authors have reported the lifetime of the state as > 10 microseconds. The work of ASARO AND PERLMAN[3] on U^{232} sources and of SCHARFF-GOLDHABER AND CO-WORKERS[4] on U^{232} and Ac^{228} sources indicates that this transition has a half life shorter than 0.2 microseconds. BOX AND KLAIBER[7] also found a pair of gamma rays in the decay of Ac^{228} differing by 58 kev (their gamma energies were 336 kev and 278 kev) which may be identical with those observed in U^{232} decay. Further discussion of Ac^{228} appears in Section 6.3.2 of Chapter 6.

A third nuclide which decays to Th^{228} is Pa^{228} as discussed in Section 8.3.4. All the radiations shown in Figure 8.20 are also observed in the decay of Pa^{228} but in addition there are many gamma rays dropping down from higher-lying levels of Th^{228} which are not reached in the alpha-decay of U^{232}.

1. P. A. Sellers, C. M. Stevens, and M. H. Studier, Phys. Rev. 94, 952 (1954).

2. J. W. Gofman and G. T. Seaborg, Paper No. 19.14, "The Transuranium Elements," National Nuclear Energy Series, Division IV, Volume 14B, Mc-Graw-Hill Book Company, Inc., New York, 1949.

3. F. Asaro and I. Perlman, Phys. Rev. 99, 37 (1955).

4. G. Scharff-Goldhaber, E. der Mateosian, G. Harbottle, and M. McKeown, Phys. Rev. 99, 180 (1955).

5. J. Kyles, C. G. Campbell, and W. J. Henderson, Proc. Phys. Soc. (London) A66, 519 (1953).

6. W. D. Brodie, Proc. Phys. Soc. (London) A67, 265 (1954).

7. H. B. Box and G. S. Klaiber, Phys. Rev. 95, 1247 (1954).

8. M. Lecoin, M. Perey, and J. Teillac, J. phys. rad. 10, 33 (1949).

8.4.6 Uranium-233. This long-lived alpha-emitter is made by the irradiation of thorium with neutrons:

$$Th^{232}(n,\gamma)Th^{233};\ Th^{233} \xrightarrow[23\ min]{\beta^-} Pa^{233} \xrightarrow[27.4\ day]{\beta^-} U^{233}$$

The half-life of U^{233} for alpha emission is 1.62×10^5 years.[1,21] It is beta stable. The discovery of U^{233} and its importance as a member of the $4n + 1$ series of radioactive isotopes is discussed in Section 7.1.4 of the preceeding chapter. Uranium-233 is of great technical importance because its high cross section for fission when irradiated with thermal neutrons and its other physical properties make it one of the few suitable materials for the industrial or military release of nuclear energy. The fission characteristics of U^{233} are thoroughly reviewed in Chapter 11.

The first careful measurements of the alpha particles of U^{233} were reported by CRANSHAW AND HARVEY[2] and by ASARO.[3] These reports have been superceded by the three studies summarized in Table 8.34. In addition to the alpha groups listed in this table TRETYAKOV AND CO-WORKERS[6] have found the following alpha groups in their alpha spectra: α_{29} (0.? %), α_{71} (0.3 %), and α_{131}(?) (0.05 %). From the study of the gamma radiations emitted by U^{233} it is quite clear that other alpha particle groups are emitted with an intensity of ~0.01 percent. RUIZ[8] for example examined the alpha spectrum in an ionization chamber alpha spectrometer operated in a gamma-alpha coincidence arrangement so that only those alpha particles in coincidence with gamma rays were recorded. This largely eliminated the interference ("tailing") from the very intense α_o group and made it possible to detect alpha transitions with 142, 210, and 318 kev less decay energy than the ground state transition. The intensity of these groups was estimated as ~0.01 percent, ~0.02 percent, and ~0.04 percent, respectively.

The intense gamma transitions which de-excite the 99 kev and 42.8 kev levels in the Th^{229} daughter nucleus have been carefully studied by many authors[9-13] of whom we cite only a few. STUDIER[11] observed gamma ray photons of 40 and 80 kev and their corresponding conversion electrons. He also reported a gamma ray of 310 kev energy in 0.1 percent intensity. BISGARD[10] found electrons corresponding to 43, 56, and 99 kev gamma transitions in coincidence with alpha particles. WEST, DAWSON, AND MANDELBERG[9] found L x-rays, a 42.8 ± 0.3 kev gamma ray and a 56.1 kev gamma ray with intensities of 4×10^{-2}, 5×10^{-4}, and 1×10^{-4} photons per alpha respectively.

Table 8.34 Alpha Groups of U^{233}

Gol'din, Novikova, Tretyakov 1955			Ruiz, Asaro, Perlman[8] 1960				Dzhelepov, Ivanov, Nedovesov[7] and Shishin 1959		
E_γ (Mev)	Energy of Th^{229} daughter level (kev)	Intensity %	E_γ	Energy of Th^{229} daughter level (kev)	Intensity %	Hindrance factor	E_γ (Mev)	Energy of Th^{229} daughter level (kev)	Intensity %
4.816	0	83.5	4.8157	0	83.7	1.17	--	0	83.4
4.773	43.2	14.9	4.773	43	14.1	3.66	--	43	14.9
4.717	100	1.6	4.719	99	1.9	10.5	--	100	1.6
4.655	164	0.07	4.652	166	0.05 ± 0.03	200	--	166 ± 3	0.06 ± 0.015
4.582	237	0.04*	not seen	237	<0.02	>100	?	234 ± 5	<0.007
4.489	333 ± 5	0.03	4.500	320	0.04 ± 0.02	10	--	316 ± 3	0.033 ± .008
							--	364 ± 5	<.005

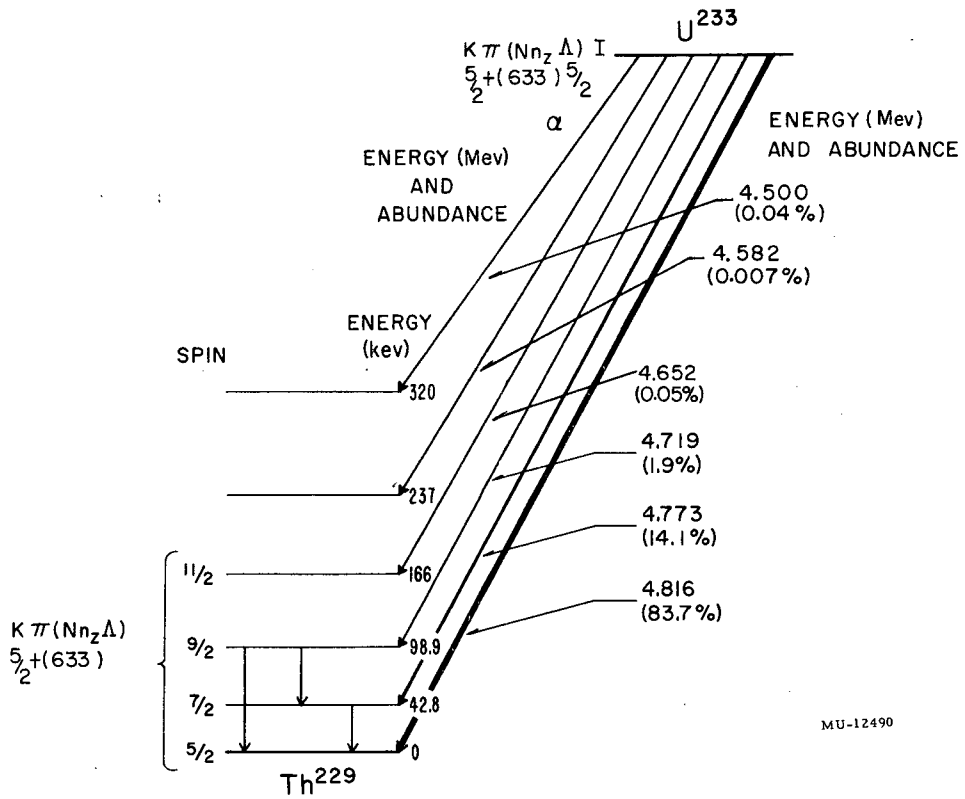
*These authors later[6] revised this intensity downward to 0.007%.

TRETYAKOV AND CO-WORKERS[6] studied conversion electron energies and assigned energies of 42.4 ± 0.2 , 54.7 ± 0.5, and 97.3 ± 0.3 kev to the three transitions under discussion. The placement of these three gamma rays in the decay scheme seems well established and is shown in Figure 8.21

We turn now to a discussion of the less intense gamma rays. The gamma transitions deduced by TRETYAKOV AND CO-WORKERS[6] from conversion electron spectra are given in Table 8.35.

RUIZ, ASARO, AND PERLMAN[8] used scintillation spectrometer techniques to study the gamma rays. In addition to an analysis of the "singles" spectrum they performed gamma-alpha, alpha-gamma, and gamma-gamma coincidence experiments which revealed the presence of photons with energies of 43, 57, 72, 94, 102, 118, 146, 164, 182, 211, 242, 282, 318, and 367 kev. The coincidence experiments suggested that a majority of these transitions terminated at the ground state of Th^{229}. These workers as well as the Russian authors cited above have formulated tentative decay schemes which include many more alpha and gamma transitions than are shown in the decay scheme given here.

Some progress has been made in the assignment of Nilsson orbitals to the odd neutron in U^{233} and in Th^{229}. The spin of U^{233} has been measured as 5/2 by interpretation of atomic spectra[14-17] and by a nuclear paramagnetic resonance study.[18] The magnetic moment is 0.51 magnetons.[18] A Nilsson state with K = 5/2 which would be a natural choice[19] for neutron number 141 is 5/2+ [633] where the brackets refer to the asymptotic quantum numbers [N n_z Λ]. The prominent alpha decay goes to the ground state of Th^{229} and to the rotational levels based on this intrinsic state. According to the concept of favored alpha decay put forth by BOHR, FROMAN, AND MOTTELSON[20] this fixes the assignment of the Th^{229} ground state also as 5/2+ [633]. This is an interesting case because usually the favored decay in an odd-A alpha emitter proceeds to an excited intrinsic state in the daughter. The fact that Th^{229} and U^{233} both have the same ground state configurations implies that the odd-particle state filled for Th^{229} is vacated when the particle becomes paired. The K = 5/2 assignment of the ground state of U^{233} has been further confirmed by the Coulombic excitation studies of NEWTON.[19] His results are summarized in Figure 8.22. The energy spacing and gamma ray pattern correspond to that expected for a 5/2, 7/2, 9/2 rotational series. Information on other intrinsic states of excitation in U^{233} comes from a study of the decay of Pa^{233} (See Section 8.3.9).



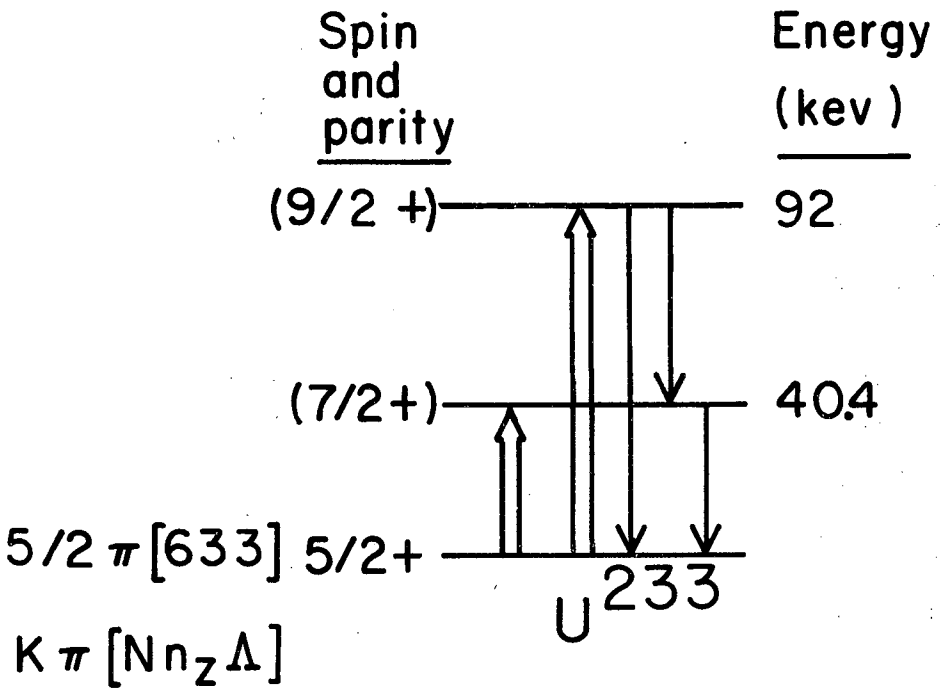
8.21 Partial decay scheme of U^{233}.

Table 8.35

Gamma Ray Transition Energies in the Decay of U^{233} from Conversion Electron Spectrum

Energy (kev)	Multipolarity
29.1 ± 0.2	M1 ?
42.4 ± 0.2	80% M1 20% E2
54.7 ± 0.5	M1 + E2
66.0 ± 1.0	
71.4 ± 0.6	
97.3 ± 0.3	E2
103.0 ± 1.0	--
121.0 ± 0.3	E2
245.3 ± 0.5	M1
248.6 ± 0.8	M1 ?
277.8 ± 1.5	M1 ?
291.5 ± 0.5	M1
317.0 ± 1.5	M1
321.0 ± 1.5	M1 ?
366.0 ± 2.0	M1 ?

From Tretyakov, Anikina, Gol'din, Novikova, and Pirogova, Soviet Physics, JETP 37, (10) 656 (1960).



8.22 Coulombic excitation of rotational levels in U^{233} (after Newton).

Several of the lowest-lying levels of Th^{229} can be interpreted as rotational levels based on the K = 5/2 ground state. The levels at 42.8 and at 98.9 kev are almost certainly the 7/2+ and 9/2+ levels, respectively of this band and the 164 kev level is probably the 11/2 member of the band. The levels at 237 kev and 320 kev were interpreted at one time[4,5] as the 13/2 and 15/2 levels of this same band but later experimental evidence[6-8] proved that these assignments could not be correct. It is clear from the gamma ray evidence[6,8] that additional intrinsic Nilsson states of Th^{229} are reached in the alpha decay of U^{233} but no very definite assignments have been made.

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1. E. K. Hyde, paper 19.15 p 1431 National Nuclear Energy Series, Volume 14B "The Transuranium Elements", edited by Seaborg, Katz, and Manning, McGraw-Hill Book Company, Inc., New York, 1949.

2. T. E. Cranshaw and J. A. Harvey, Can. J. Research 26A, 243 (1948).

3. F. Asaro, University of California Radiation Laboratory Report, UCRL-3180, June (1953).

4. L. L. Gol'din, E. F. Tretyakov, and G. I. Novikova, Proceedings of a Conference of the Academy of Sciences of the USSR on the Peaceful Uses of Atomic Energy, July 1955; English translation available from Superintendent of Documents, U. S. Government Printing Office, Washington 25, D. C.

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8. C. Ruiz, F. Asaro, and I. Perlman, unpublished results (1960); see C. Ruiz, Ph.D. Thesis, University of California Radiation Laboratory Report, UCRL- (1961).

9. D. West, J. K. Dawson, and C. J. Mandleberg, Phil. Mag. 43, 875 (1952).

10. K. M. Bisgard, Proc. Phys. Soc. (London) 65A, 677 (1952).

11. M. H. Studier, paper 1.3 in Report TID-5223 "Production and Separation of U^{233}"; see also report AECD-2444.

12. D. C. Dunlavey, University of California Radiation Laboratory Report, UCRL-1911, August 1952.

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14. K. L. Vander Sluis and J. R. McNally, Jr., J. Opt. Soc. Amer. 44, 87 (1954); 45, 65 (1955).

15. N. I. Kaliteevskii and M. P. Chaika, Doklady, Akad. Nauk SSSR 103, 49 (1955); Optika i Specktroskopiya 1, 809 (1956); AEC-tr-2890.

16. L. A. Korostyleva, A. R. Striganov, and N. M. Iashin, Soviet Physics, JETP 1, 310 (1955).

17. A. G. Zimin and N. M. Yashin, Doklady, Akad. Nauk SSSR 109, 283 (1956); Soviet Physics, Doklady 1, 419 (1957).

18. P. B. Dorain, C. A. Hutchison, Jr., and E. Wong, Phys. Rev. 105, 1307 (1957).

19. J. O. Newton, Nuclear Physics 5, 218 (1957).

20. Bohr, Fröman, and Mottelson, Kgl. Danske Videnskab. Selskab. Mat. Fys. Medd. 29, No. 10 (1955).

21. Dokuchayev and Osipov, Atomnaya Energ. 6, 73 (1959).

8.4.8 Uranium-234. This isotope appears in the natural uranium decay chain in which context it has traditionally borne the name Uranium-II. Its position as a member of the uranium series is discussed in Chapter 6. It is a beta-stable alpha-emitter with a half life of 2.48×10^5 years. (Refer to Table 6.2 in Section 6.2 of Chapter 6.) The alpha particle energy was reported by HARVEY, JACKSON, EASTWOOD, AND HANNA[1] to be 4.768 Mev on the basis of careful ionization chamber measurements. GOL'DIN, NOVIKOVA, AND TRETYAKOV[2] measured the alpha spectrum in an enriched sample of 7 percent abundance and found two groups; the main group of energy of 4.768 ± .001 Mev energy was in 72 percent abundance; a second group 51.5 kev lower in energy was in 28 percent abundance. Additional studies with more highly-enriched U^{234} should show the additional alpha group of 4.59 Mev energy and lower intensity which is required by the gamma spectrum.

TEILLAC[5] saw the conversion electrons of a gamma ray of about 50 kev energy. BELL AND CO-WORKERS[6] reported gamma rays of 53 and 118 kev as well as K x-rays from the conversion of the 118 kev gamma ray. VALLADAS[3] performed coincidence experiments between alpha particles and gamma rays and concluded that there were at least three alpha groups in the U^{234} spectrum with the following abundances: α_0 (77 percent), α_{56} (23 percent), and α_{170} (~0.3 percent). SCHARFF-GOLDHABER[4] also reports the presence of the gamma rays of 50 and 117 kev energy.

These observations are summarized in the decay scheme of Figure 8.23. This is a typical spectrum for an even-even alpha-emitter in the heavy element region. There seems little doubt that the observed alpha groups populate three members of the ground state rotational band of Th^{234} (ionium) and that the 4+ level is de-excited with the usual cascade of electric quadrupole transitions, the second of which is very highly converted. Presumably if highly enriched samples of U^{234} were studied more carefully some alpha decay to higher-lying members of the even parity rotational band and to a 1- level would also be found but in quite low intensity.

Information on other levels of Th^{230} has been obtained from the gamma radiations seen in the electron capture branch of the decay of Pa^{230} (See Section 7.3.1 of the preceeding chapter).

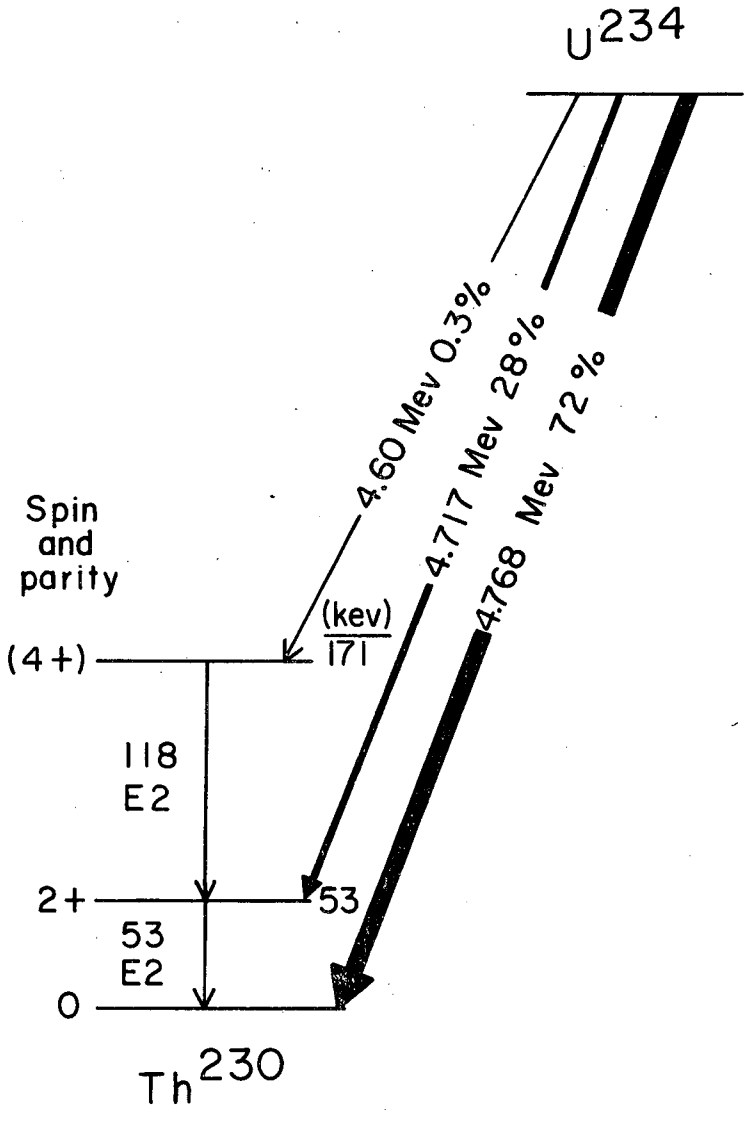

Fig. 8.23 Decay scheme of U^{234}.

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2. L. I. Gol'din, E. F. Novikova, G. I. Tretyakov, Conference of the Academy of Sciences of the USSR on the Peaceful Uses of Atomic Energy. Session of the Division of Physical and Mathematical Sciences, Moscow, July, 1955. English translation available from Superintendent of Documents, U. S. Government Printing Office, Washington 25, D. C.

3. G. Valladas, Compt. rend. 237, 1673 (1953).

4. G. Scharff-Goldhaber, reported in Brookhaven National Laboratory Report BNL-103, June, 1951 (unpublished).

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reported in Oak Ridge National Laboratory Report, ORNL-1164, April, 1952 (unpublished).

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Physica 21, 719 (1955).

8.4.9. Uranium-235.‡ All uranium minerals contain U^{235} in 0.720 percent isotopic abundance. Uranium-235 is the parent activity of the actino-uranium family of natural radioactivities. This series is thoroughly discussed in Section 6.4 of Chapter 6. The fission characteristics of U^{235} are thoroughly discussed in Chapter 11.

Uranium-235 is an alpha-emitter with a half life of 7.13×10^8 years. The low specific activity of 4.5 disintegrations per minute per microgram causes considerable difficulty in the determination of the alpha spectrum. A number of analyses of this spectrum have been carried out by absorption or ionization chamber techniques[1] but these have been superceded by the work of PILGER, ASARO, AND PERLMAN.[2,3] These authors exposed a weak sample of U^{235} for two weeks in a double-focusing magnetic spectrometer of high resolution and moderate transmission (4×10^{-4} of 4π). The resulting spectrum is shown in Figure 8.24. The peak at 4.354 Mev is complex and consists of three or more alpha groups. The analysis of this complex peak into its several components is somewhat uncertain. The alpha groups are summarized in Table 8.35.

The gamma rays associated with the decay of U^{235} have been studied by a number of groups; among recently published studies are those of PILGER, ASARO, STEPHENS, AND PERLMAN,[4] of MALICH[5] of JOHANSSON[6] and of STEPHENS[7] and of FILIMONOV AND PSHENICHNIKOV.[13] Gamma rays have been identified at: 95 kev (K x-rays) (9%), 110 kev (5%), 144 kev (12%), ~165 kev ($\leq$ 4%), 185 kev (55%), and 205 kev (~4%). Other gamma rays have been reported but the data are not so conclusive as for those mentioned above. The low specific activity of U^{235} makes it quite difficult to study the conversion electrons. Coincidence studies have established that the 204 kev gamma ray is in coincidence with both the 110 and 185 kev gamma rays. STROMINGER[14] has measured the half-life of the 186 kev E1 transition and reports the value $(0.77 \pm 0.12) \times 10^{-9}$ seconds. This transition is slower by a factor of 6×10^4 than the single proton lifetime prediction. This hindrance can be understood qualitatively in terms of

‡ Some recent Russian work on the alpha spectrum of U^{235} indicates a somewhat different resolution of the close-lying groups than is shown in 8.25. This information was received too late for inclusion in this report.

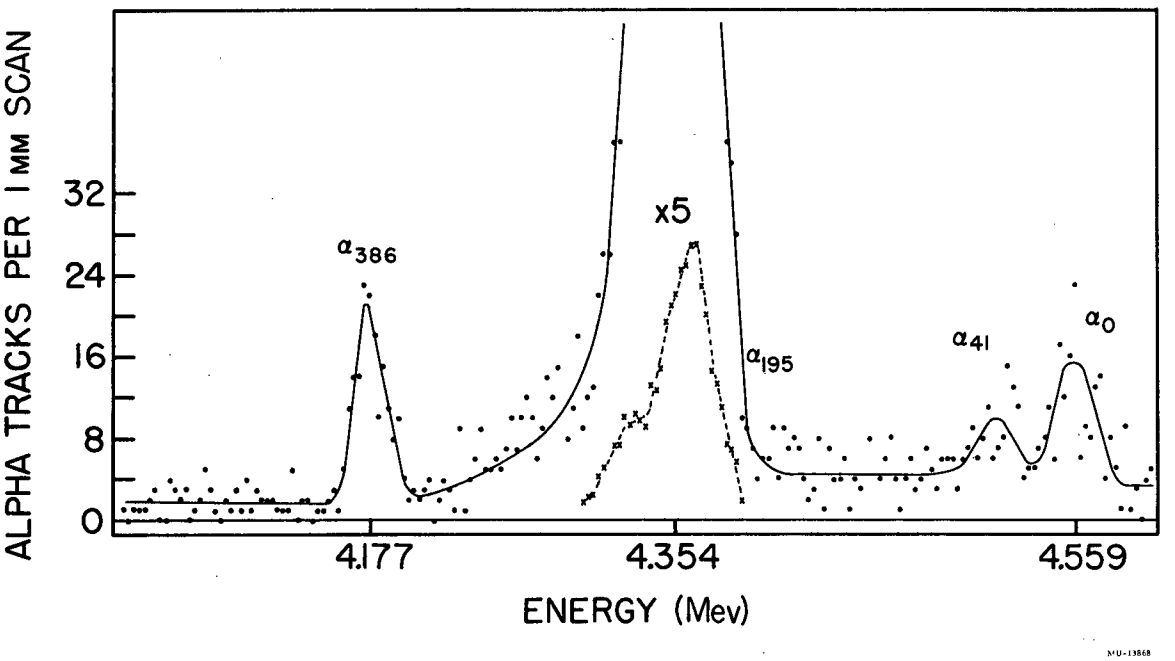

Fig. 8.24 Alpha particle spectrum of U^{235} taken by Pilger, Asaro, and Perlman.[3] The full width of the peaks at half their maximum height is 15-20 kev. The peak at 4.354 Mev is complex and consists of three or more alpha groups. Other resolutions would be consistent with the data. The alpha-disintegration rate of the sample was 480 per minute, of which 67 percent belonged to U^{235}. The rest belonged to U^{234} which served as a convenient energy standard. The exposure time was 13 days.

8.4.10 Uranium-235m. The primary product of the alpha decay of Pu^{239} is an isomeric form of U^{235} with a half-life of 26.5 minutes. This isomer lies less than 100 electron volts above the ground state and decays to the ground state by an E3 gamma transition. The formation and decay of U^{235m} are thoroughly discussed in connection with the alpha decay of Pu^{239}. See Section 9.2.9.

Table 8.35

Alpha Groups of U^{235}

(Pilger, Stephens, Asaro, and Perlman[3])

Particle energy (Mev)			
4.559 ± 0.0.5	6.7	0	960
4.520	2.7	40 (42)	1200
4.466 (?)[b]	0.9	95 ± 10	~1500
4.369	29	191 (185)	9.5
4.349	43	204	4.5
4.327	12	224	11
4.177 ± 0.004[a]	5.8	390	1.5

Decay Energy (Q_α) 4.639 ± 0.015 Mev

[a]Relative to U^{234}, reported as 4.768 ± 0.002 relative to Po^{210}.

[b]The existence of this group is regarded as questionable.

a violation of selection rules in the asymptotic quantum numbers. Such delays are a general phenomenon for E1 transitions in the heavy element region as discussed in Section 3.5.7 of Chapter 3.

The alpha and gamma radiation data have been combined by PILGER, STEPHENS, ASARO, AND PERLMAN[3] into the decay scheme which is shown here as Figure 8.25. In this scheme the data have been interpreted in terms of three rotational bands having base states at 0, 185, and 389 kev. Specific assignments of NILSSON states have been made for grounds which will be briefly discussed.

The ground state of U^{235} has a measured spin of 7/2[8,8a] and has been given the Nilsson assignment 7/2- [743] by NEWTON,[9] ASARO AND PERLMAN[10] and others. This assignment can be considered quite well established. It is supported by the Coulombic excitation studies of NEWTON[9] who excited the 9/2 and 11/2 rotational levels and by the alpha decay of Pu^{239} which takes place predominantly to a K = 1/2 intrinsic state of U^{235} which lies within 0.1 electron volts of the ground state. This state decays to the ground state by an E3 transition with a half life of 26 minutes. This evidence is discussed in connection with the alpha decay of Pu^{239} in Section 9.2.9 of the next chapter.

The 4.117 Mev alpha particle emission is unhindered. According to the concept of favored alpha transitions formulated by BOHR, FRÖMAN, AND MOTTELSON[11] unhindered alpha decay in odd-A alpha emitters proceeds to a state in the daughter nucleus with the same intrinsic state assignment as the parent. On this basis the 390 kev level in Th^{231} would also be given the assignment 7/2- [743]. This level decays to several members of the group of levels around 185 kev by transitions which appear to be predominantly M1.[7] Furthermore, the low alpha hindrance factor to the levels around 185 kev suggests that these levels are rather similar to the 7/2- [743] state. Consideration of the Nilsson diagram shows that by far the most reasonable assignment for the 185 kev level is 5/2- [752]. The unusually close spacing of the rotational band based on this level will be discussed presently. The ground and 40 kev levels of Th^{231} are connected with the 185 kev level by E1 transitions,[7] showing that the parity of these levels is positive. It seems quite likely that these levels are members of the same rotational band, and, if this is the case, the above-mentioned E1 transitions permit only spins of 3/2 or 5/2 for the ground state of Th^{231}. The 40 kev spacing of these two levels suggests that the spin is 5/2 rather than 3/2, since the rotational constant, $\frac{\hbar^2}{2\mathcal{I}}$, derived from the

5/2 spin is in better agreement with those for other odd-mass nuclei in this region. The Nilsson assignment for the ground state of Th^{231} is thus very likely 5/2+ [633]. This would be expected since the ground state of U^{233}, which has the same number of neutrons, has been given this assignment. Such an assignment also fits in well with the decay scheme of Th^{231} discussed in Section 8.2.9.

The main features of the decay scheme shown in Figure 8.25 have now been developed, and there remains only the problem of the very close spacing of the 5/2- [752] band. This can be explained as due to the interaction between the 7/2- [743] and 5/2- [752] rotational bands which is brought about by the Coriolis forces. This interaction has been discussed by KERMAN.[12] The effect is unusually large in this case for two reasons. First, the levels involved are both derived from the j 15/2 single particle level; and second, these two bands in Th^{231} are unusually close together because the ground state of Th^{231} (5/2+ [633]) lies between them on the Nilsson diagram. Calculations indicate that a spacing as close as that shown in Figure 8.25 is not at all unreasonable in view of the above considerations. It should be added here that the resolution of the complex alpha group leading to this band is not completely certain. The dashed levels in Figure 8.25 represents a resolution consistent with both the experimental data and the above interpretation.

The large Coriolis interaction in Th^{231} is a relatively new consideration in analyzing decay schemes, and thus it should still be considered somewhat tentative. On the other hand, no other explanation of the levels of Th^{231} fits the data presently available so well.

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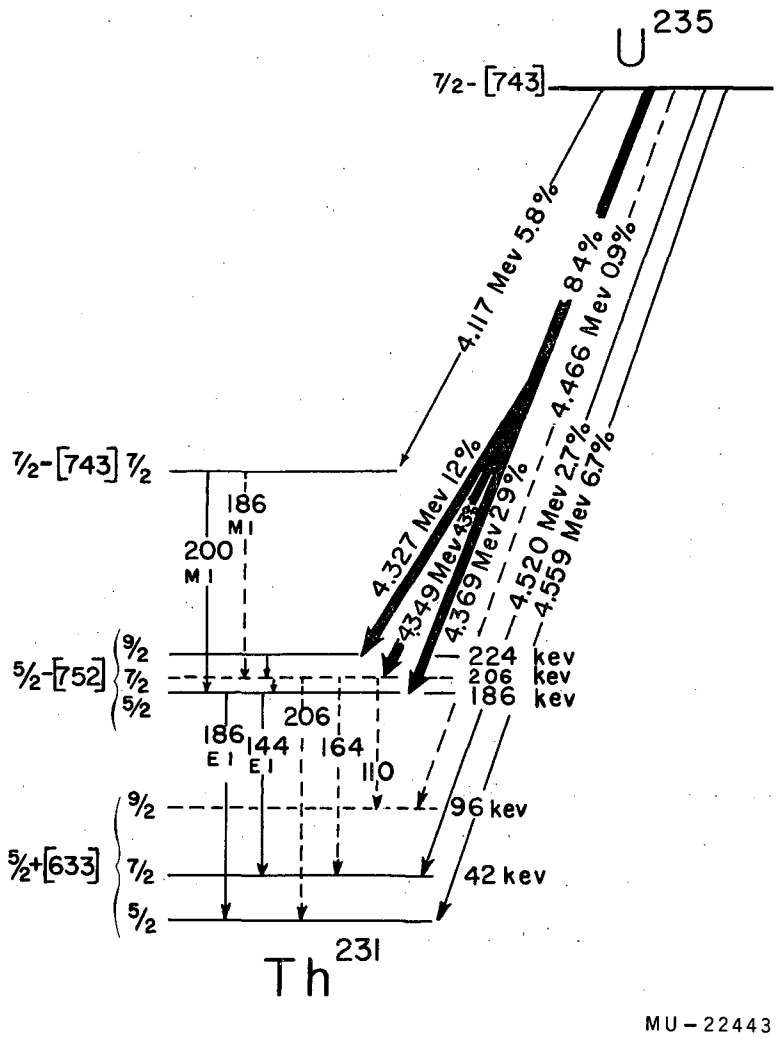

Fig. 8.25 Decay scheme of U^{235} as formulated by Pilger, Asaro, Stephens, and Perlman. The Nilsson state assignments are believed to be firm but the exact position and alpha population of the rotational band around 186 kev are uncertain.

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8a. C. A. Hutchison, Jr., P. M. Llewellyn, E. Wong, P. Dorain, Phys. Rev. 102, 292 (1956).

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12. A. K. Kerman, Dan. Mat. Fys. Medd. 30, no. 15 (1956).

13. Yu. I. Filimonov and B. V. Pshenichnikov, Zhur. Eksptl. i Teoret. Fiz. 35, 548 (1958).

14. D. Strominger, Phys. Rev. 114, 502 (1959).

8.4.11 Uranium-236. WILLIAMS AND YUSTER[1] first detected U^{236} by mass spectrographic analysis of an enriched sample of U^{235} which had been irradiated with slow neutrons. GHIORSO, BRITTAIN, MANNING, AND SEABORG[2] first detected the alpha particles which U^{236} was expected to emit, and set a value of about 2×10^7 years for the half-life. Uranium-236 is beta stable. FLEMING, GHIORSO, AND CUNNINGHAM[3] measured the specific activity of U^{236} samples which had been enriched to 97 percent isotopic purity by electromagnetic separation. The specific activity is $(1.406 \pm 0.011) \times 10^5$ disintegrations per minute per milligram corresponding to a half-life of $(2.391 \pm 0.018) \times 10^7$ years. This value supercedes an earlier determination which yielded the value 2.457×10^7 years.[4] The half-life of U^{236} is too short for any U^{236} to have survived since the formation of the elements but it is long enough that U^{236} may have contributed significantly to the heating of the earth during its early history. ROSENBLATT[5] has put forward this suggestion, but KOHMAN[6] is inclined to discount it.

The energy of the main alpha particle group has been measured[2] by the ion chamber method to be 4.499 ± 0.004 Mev. Evidence for other groups has been found indirectly. DUNLAVEY AND SEABORG[7] impregnated photographic emulsions with U^{236} and found conversion electrons from a ~50-kev gamma ray in coincidence with 27 percent of the alpha particles. GROVER AND SEABORG[8] found a gamma ray of 163 kev energy in ~0.5 percent abundance. From these results, the principal alpha groups are believed to be the following:

4.499 Mev (73 percent), 4.45 Mev (27 percent),
and 4.337 Mev (~0.5 percent).

Further work on U^{236} may be expected to show that the decay scheme of U^{236} is closely similar to those of all even-even alpha emitters in this mass region.≠

Further information on the excited levels of Th^{232} has been obtained from the excitation of nuclear states by the Coulombic excitation method using charged particle beams of energy below the potential barrier for nuclear reactions. The first excited 2+ level at 53 kev was produced by several investigating teams.[9-12] STEPHENS, DIAMOND, AND PERLMAN[13] bombarded thorium targets with argon ions of high energy but of insufficient energy to

≠ KOMAR, KOROLEV AND KOCHAROV[16] report the following alpha groups: α_0 (74%) α_{50} (26%) and α_{160} (0.26%). They give 4.488 ± .003 Mev as the energy of the ground state transition.

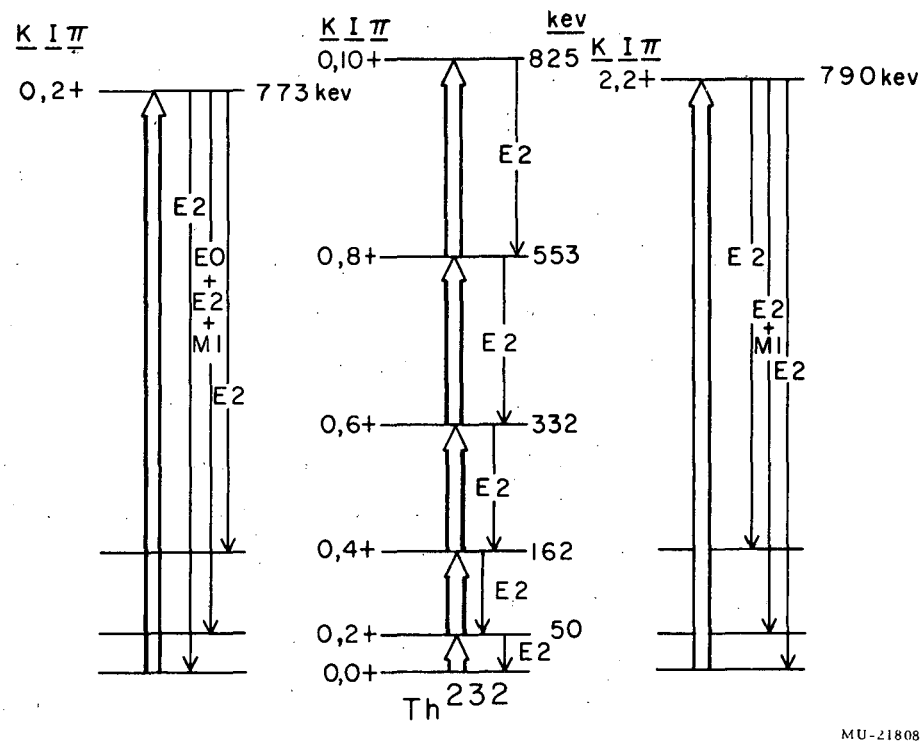

Fig. 8.26 Levels of Th^{232} revealed by the Coulombic excitation process. The energy of each level is given in kev at the right of the level.

penetrate the Coulomb barrier and were able to excite several levels of the ground state rotational band up to a 10+ level at 825 kev. These results are summarized in Figure 8.26. STELSON AND McGOWAN[11,12] were also able to excite a 2+ state at 790 kev which decays by E2 radiation to the 0+, and 2+ members of the ground state rotational band. This level at 790 kev is probably the first γ-vibrational state of the collective excitation of the Th^{232} nucleus. DURHAM, RESTER, AND CLASS confirmed the excitation of the 790 kev level and its assignment as a state of vibrational excitation. In addition they reported the excitation of a level at 773 kev which they assigned to the 2+ member of the beta-vibrational band. An interesting feature of this study was the presence of prominent conversion electrons of a 723 kev transition connecting the vibrational level at 773 with the 2+ level of the ground state rotational band at 50 kev. The prominent conversion electrons arise from a strong E0 component in this gamma transition. These results were confirmed by DIAMOND AND ELBEK. These last-named authors also have unpublished data on the excitation of states of negative parity by the Coulomb excitation process. These lie about 1 Mev above ground. The well-established levels of Th^{232} are summarized in Figure 8.26.

The chief method of preparation of U^{236} is by the neutron capture reaction on U^{235} for which the cross-section is 107 barns. Samples of high isotopic purity are prepared by the intense neutron irradiation of U^{235} followed by electromagnetic separation. The formation of U^{236} by the reaction U^{235} (n,γ) U^{236} is an important factor in the operation of nuclear reactors burning U^{235} not only because of the change in isotopic composition of the fuel but because U^{236} is an intermediate in the production of other heavy element nuclides such as Np^{237}.

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16. A. P. Komar, G. A. Korolev and G. E. Kocharov, Sov. Phys., JETP, 11, 1038 (1960).

8.4.12 Uranium-237. Uranium-237 is a beta emitter with a half life of 6.75 days. This nuclide is a collateral member of the 4n + 1 series by virtue of its decay into Np^{237}. The discovery of U^{237} is discussed in connection with the discovery of Np^{237} in Section 7.1 of the preceeding chapter.

Uranium-237 is usually prepared by fast neutron bombardment of U^{238} by the reaction:

$$U^{238}\ (n,2n)\ U^{237}$$

It is a prominent component in the radioactive debris from the testing of nuclear explosive devices in which an intense fast neutron flux is generated in the vicinity of U^{238}. The nuclide can also be prepared by the following cyclotron reactions:

$$U^{238}\ (d,t)\ U^{237}$$

$$U^{238}\ (\alpha,\alpha n)\ U^{237}$$

$$U^{238}\ (p,pn)\ U^{237}$$

We turn now to a detailed review of its complex radiations.

Of those nuclei in the heavy-element region whose spectroscopic properties have been widely studied, few are as interesting as Np^{237} the daughter of U^{237}; its levels have provided a wealth of information with which to test the Bohr-Mottelson theory, both in its general and in its more detailed aspects. The Np^{237} levels are populated by the alpha decay of Am^{241}, by the beta decay of U^{237}, and by the electron capture decay of Pu^{237}; this section on U^{237} decay will supplement the more detailed discussion presented in the Section 9.3.6 covering Am^{241} decay. The study of U^{237} neatly supplements the work on Am^{241} since the decay of the former populates chiefly some high-lying levels of Np^{237} whereas the alpha decay leads overwhelmingly to the low-lying levels.

Spectroscopic studies of U^{237} decay have been carried out by WAGNER, FREEDMAN, ENGELKEMEIR, AND HUIZENGA,[1] BARANOV AND SHLYAGIN,[2] BUNKER, MIZE, AND STARNER,[3] and by RASMUSSEN, CANAVAN, AND HOLLANDER.[4] Figure 8.27 shows the Np^{237} energy level scheme as summarized by RASMUSSEN, CANAVAN, AND HOLLANDER,[4] which includes both the Am^{241} and U^{237} information. The discussion here will follow closely that given in their paper.

The principal electromagnetic radiations observed in the U^{237} spectrum are photons of 60-kev (38%), K x-rays (55%), 163-kev (3.6%), 208-kev (24%), 266-kev (0.9%), 332-kev (~2%), and 365-kev (~0.1%); the 60-kev

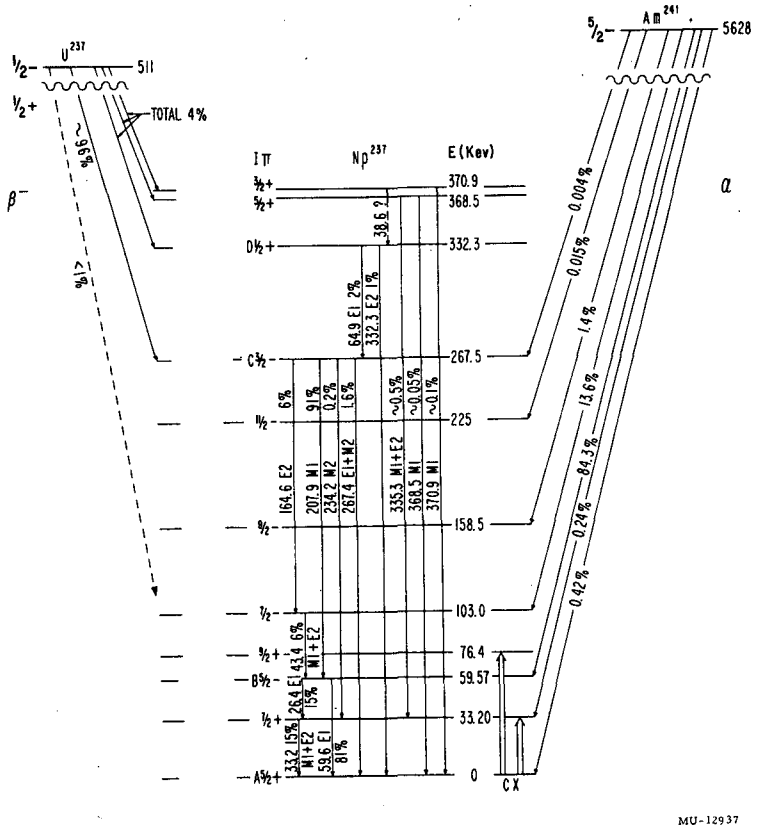

Fig. 8.27 Decay scheme of U^{237}. The alpha transitions of Am^{241} to the same daughter nucleus shown for comparison. Note that U^{237} populates primarily the level at 267.5 kev. Transition energies, multipolarities, and gross abundances are shown. Nilsson state assignments are given for intrinsic particle states. The levels at 225 and 158.5 kev which appear in Am^{241} alpha decay and the level at 76.4 kev excited coulombically (symbol CX) are not observed in the decay of U^{237}.

Table 8.36

Gamma transitions of U^{237}

Energy (kev)	Multipolarity assignment*	Total estimated transition intensity
26.35	E1	15
33.20	M1 + E2	15
43.46	M1 + E2	7
59.57	E1	81
64.8	E2	3
113.9		
164.6	E2	7
207.9	M1	89
234.2	M2	~0.2
267.5	E1 + M2	1.7
332.3	E2	1.5
335.3	M1 + E2	~0.2
368.5	M1	~0.05
370.9	M1	~0.10

*Chiefly from L-subshell conversion ratios.

photon is familiar because of its prominent position in the decay of Am^{241}. The other transitions shown in Figure 8.27 and listed in Table 8.36 are either highly converted or too weak to appear in the scintillation spectrum. These are discussed thoroughly by RASMUSSEN et al.[4] The end point of the main spectrum is 248 kev.[1,2,4] A low limit of 0.1 percent has been set on beta particles leading directly to the ground state or low-lying levels of Np^{237}. The chief beta decay goes almost entirely (~96 percent) to the 267.5 kev level but minor branching to higher-lying states must occur.

The level at 267.5 kev de-excites by four transitions, all of different multipole orders (a most unusual and interesting situation). Of these, the 207.9 kev magnetic dipole transition is the most prominent. From our knowledge of the spins and parities of the lower states in Np^{237} (from Am^{241} decay) and from the multipolarity information on the transitions which de-excite the 267.5 kev state, the spin and parity assignments of this state are 3/2-.

The eight levels populated directly or indirectly by the beta decay of U^{237} can be grouped into four rotational bands; these are displayed schematically in Figure 8.28. Band A, based on the ground state, has K = 5/2 with positive parity; Band B based on the 59.6 kev level has K = 5/2 with negative parity; Band C with one known level at 267.5 kev has K = 3/2 with negative parity; Band D based on the 332.3 kev level has K = 1/2 with positive parity. Energies of levels in a K = 1/2 type band are given by the following formula (See Chapter 3):

$$E_I = \frac{\hbar^2}{2\Im} \left[I\,(I+1) + a\,(-)^{I+1/2}\,(I+1/2) \right]$$

Using the three level energies, 332.3, 368.5, and 370.9 kev and the spins 1/2, 3/2, and 5/2 respectively, one calculates a value of the splitting constant, $\hbar^2/2\Im$ = 6.2 kev which compares closely with that of Band B. One cannot, however, in this case test the accuracy with which the equation fits the experimental energy ratios, because knowledge of just two excited states of a K = 1/2 rotational band serves only to define the two parameters in the equation.

The experimental levels of Np^{237} provide some interesting comparisons with the "branching-ratio rule" of ALAGA, ALDER, BOHR, AND MOTTELSON.[5] This rule states that in photon transitions of a single multipole type which originate at a common level and terminate at the various levels of a rotational band,

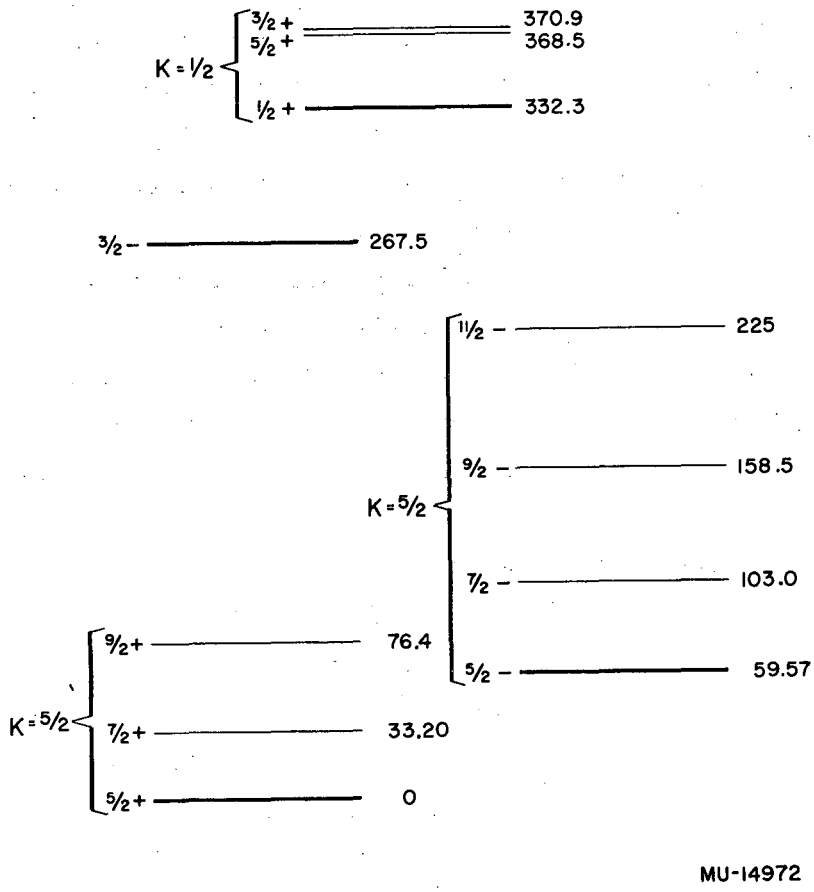

Fig. 8.28 Levels of the Np^{237} nucleus grouped as rotational bands. Each group of levels represents collective rotational excitation of a base state which is an eigenstate of the odd proton in an anisotropic harmonic oscillator potential. Levels here shown are seen in the decay of Np^{237} and Am^{241} or both. The only levels not seen in the decay of Np^{237} are the levels at 225, 158.5, and 387.5 kev.

the intrinsic nuclear transition probabilities are identical and observable differences in the photon intensities should result only from the different energies and angular momenta involved; both are independent effects and easily corrected for,

Two pairs of gamma-rays may be compared in the Np^{237} levels; these are the 26.4 - 59.6 kev pair of electric dipole transitions, and the electric quadrupole components of the 164.6 and 207.9 kev photons. The agreement with theory in both cases is poor, and in the latter pair there is at least a factor of ten discrepancy. The key to understanding the lack of agreement lies in the recognition that these cases involve transitions much retarded from the single-particle rates. The disagreement is interpreted within the framework of the Bohr-Mottelson theory as an indication that those components of initial or final wave functions actually contributing to the transition matrix element have a different K value from the main components of the wave function. In general, such disagreement is found in abnormally retarded transitions; those transitions proceeding at nearly the single particle rate generally show good agreement with the branching ratio rules.

Explanation of the factors causing the retardation of the transitions mentioned here has been advanced in terms of selection rules involving the Nilsson eigenfunctions (states) appropriate to these highly deformed nuclei, represented by an asymptotic limit in the theory. The low energy E1 transitions all violate these selection rules, as do also the E2 transitions mentioned above. In addition, very strong M1 radiation from the 267.5 kev state (207.9 kev) is retarded by more than 10^4 from the single particle rate, and this transition also violates the "asymptotic" selection rules.

There are interesting cases in Np^{237} of anomalous electric dipole conversion coefficients, a significant phenomenon which has yet to receive adequate theoretical explanation. The retarded 60 kev transition to ground has a normal L_{III} conversion coefficient but abnormally high values for conversion in the L_I and L_{II} subshells. The retarded 267.5 kev transition appears to have a high K-conversion coefficient. However, in the same nucleus, the 65-kev transition from the 322-kev level seems to have normal conversion coefficients. Anomalies due to dynamic nuclear structure effects can arise by virtue of the fact that different nuclear matrix elements can produce internal conversion when electron density lies within the nuclear volume. The anomalous contributions are observable only when the normal

radiative process is highly retarded and the "anomalous" conversion process is not, and it is consistent with this interpretation that in the case of electric dipole anomalies only the penetrating electrons ($s_{1/2}$ and $p_{1/2}$) show definite anomalies.

For further details on the radiations of U^{237} or the interpretation of the U^{237} decay scheme the reader is referred to the comprehensive paper of RASMUSSEN, CANAVAN, AND HOLLANDER.[4]

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8.4.13 Uranium-238. This is the chief constituent of natural uranium and the progenitor of the uranium series of radioelements. Its properties are fully discussed in Section 6.2 of Chapter 6.

The decay scheme of U^{238} is very simple and can be briefly recapitulated here. Uranium-238 is beta stable. It's half-life for alpha decay is $(4.51 \pm 0.01) \times 10^9$ years corresponding to a specific activity of 738.6 ± 1.6 disintegrations per minute per milligram. The energy of the main group of alpha particles, emitted in 77 percent abundance is 4.195 Mev. Conversion electrons of a 48 kev gamma ray have been observed indicating that the alpha decay is complex; an alpha group 48 kev lower in energy than the main group is present in 23 percent abundance. This 48 kev state in the daughter Th^{234} is undoubtedly the (2+) member of the ground state rotational band. Uranium-238 very probably undergoes slight alpha branching decay to higher-lying members of the ground state rotational band but this branching has not been observed because of experimental difficulties stemming from the low specific activity of U^{238}.

8.4.14 Uranium-239. In 1937 HAHN, MEITNER, AND STRASSMAN[1] noted that a new isotope of uranium with a 23-minute half-life was produced when natural uranium was irradiated with slow neutrons. They correctly identified this activity as U^{239}. The decay of U^{239} produces Np^{239}, the first form of element 93 to be identified. The discovery of neptunium via the reactions:

$$U^{238}\ (n,\gamma)\ U^{239}\ ;\ U^{239} \xrightarrow[23\ min]{\beta^-} Np^{239}$$

is treated in Section 9.1.11 of Chapter 9.

Uranium-239 emits beta particles with an end-point energy of 1.21 Mev.[2-4] A single gamma ray of 73.6 kev has been observed.[2-4] The isotope Am^{243} also decays to Np^{239} and additional information on the excited levels of Np^{239} comes from studies of the gamma radiations accompanying Am^{243} decay. Refer to Section 9.1.11, Chapter 9.

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8.4.15 Uranium-240. The isotope U^{240} was found by HYDE, STUDIER, AND MANNING[1] in uranium samples which had been irradiated with a high flux of neutrons.

$$U^{238}\ (n,\gamma)\ U^{239}\ (n,\gamma)\ U^{240}$$

This isotope decays with a half life of 14 hours to a 7.3 minute isomer of Np^{240}. The most careful study of the radiations of U^{240} and its daughter was made by BUNKER, DROPESKY, KNIGHT, STARNER, AND WARREN[2] whose findings are summarized in the section of Chapter 9 (Section 9.1.12) dealing with Np^{240}. The decay scheme of U^{240} is very simple and the published information on it is summarized in the accompanying figure (Figure 8.29).

Uranium-240 has also been isolated as the daughter of Pu^{244}.

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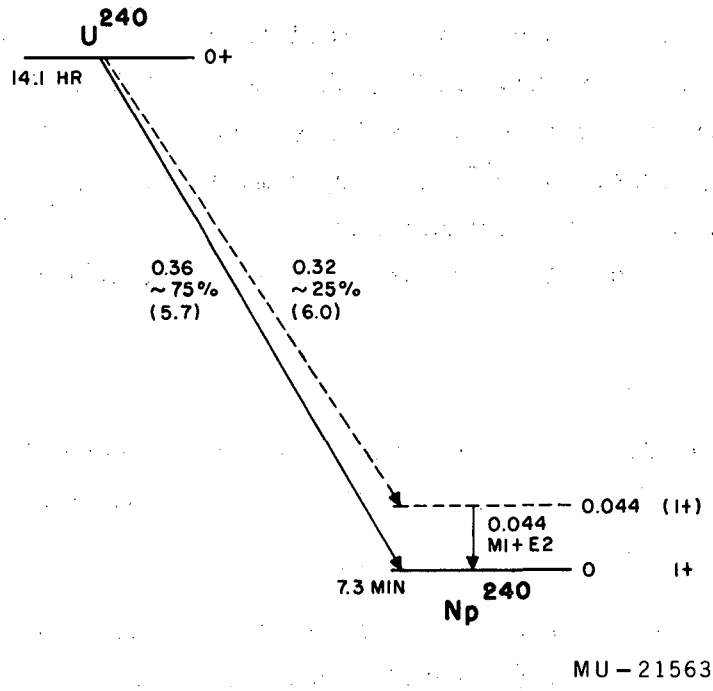

Fig. 8.29 Decay scheme of U^{240} as drawn by Bunker and co-workers.

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