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Abstract

The gamma-ray spectrum of 2.3-day Np^{239} has been studied with high-resolution lithium-drifted germanium spectrometers, and twelve new gamma rays of low intensity have been identified. Ten of these are characterized as the de-excitation spectrum of a K = 1/2 band based at 469.8 keV in Pu^{239}. The decoupling parameter (a) of this band is found to be 0.48. A comparison is made of the experimental and theoretical photon relative transition intensities and reasonable agreement is obtained for the case: $K_i = 1/2$, $b_{E1} = 0$. The properties of this band are consistent with those expected for Nilsson state 1/2- [501]. Estimates of the log ft values of Np^{239} beta decay to levels of the new band are made and are discussed in terms of the states involved.

1. Introduction

The energy levels of $_{94}Pu^{239}$ have been studied extensively from the beta decay of $_{93}Np^{239}$ [1,2]) and $_{95}Am^{239}$ [3]) and from the alpha decay of $_{96}Cm^{243}$ [4]). These levels have been characterized in terms of four intrinsic Nilsson states and the rotational excitations of these states. These Nilsson states and their energies are: 1/2+ [631], ground; 5/2+ [622], 285.5 keV; 7/2- [743], 391.6 keV; and 7/2+ [624] or 5/2+ [633], 511.9 keV. Two very weak gamma-rays of ~440 and ~490 keV were found in the scintillation spectrum of Np^{239} by Lefevre, Kinderman, and Van Tuyl[5]), and these have not been placed in the Pu^{239} level scheme. In an attempt to gain further information about these and other possible weak radiations in Pu^{239}, we have examined the photon spectrum of Np^{239} with high-resolution lithium-drifted germanium detectors. The results of this examination are reported here.

Considerable structure is found in the energy region of 400-500 keV, and twelve new gamma-rays are definitely assigned to the decay of Np^{239}. From energy sum and difference relationships among these gamma-rays a new set of levels in Pu^{239} is defined that appears to have the properties of a K = 1/2 band. Assignment of this band is made to the Nilsson state 1/2- [501]. Two of the new transitions are assigned to the de-excitation of the previously known 511.9 keV level.

2. Preparation of Source

The source of Np^{239} used in this study was prepared by irradiation of several grams of highly enriched U^{238} in the LPTR reactor at Livermore, California for 36 hours at an average flux of 5.4×10^{12} neutrons cm^{-2} sec^{-1}. The following chemical procedure was utilized for purification of the Np^{239}

from all other activities, including fission products: The uranium target was dissolved in concentrated hydrochloric acid with addition of a minimum amount of nitric acid, and carrier solutions of barium, strontium, cadmium, and zirconium were added. After reduction of neptunium to the tetrapositive state by addition of hydrazine and ferrous ion, phosphoric acid was added to precipitate $Zr_3(PO_4)_4$, which carried the neptunium activity. The precipitate was dissolved in 1 M HNO_3 - 4M HF and further purification of neptunium was made by co-precipitating the activity on lanthanum fluoride and lanthanum hydroxide, with thorough washing of each precipitate. The final lanthanum hydroxide precipitate was dissolved in concentrated hydrochloric acid, and from this solution the neptunium activity was adsorbed on to a 3 mm × 5 cm Dowex-A2 anion exchange column. The column was "washed" with solutions of 6 M and 4 M hydrochloric acid to remove remaining impurities (e.g., Zr, Nb) and the neptunium was finally eluted with 0.1 M hydrochloric acid. This solution was evaporated for gamma counting.

3. Experimental Apparatus

The gamma spectrum was studied with two lithium-drifted germanium detector assemblies[6]), coupled to low-noise preamplifier-amplifiers designed by Goulding and Landis[7,8]) and fabricated at this Laboratory[9]). Pulse-height analyses were made with 400-channel R.I.D.L. analyzers. Dimensions of the detectors are 2 cm^2 × 7 mm deep and 6 cm^2 × 9 mm deep, respectively. The larger, more efficient, detector was used for the first scans of the spectrum and for measuring the half-lives of the gamma lines. The smaller detector with better resolution (half-width 2.3 keV at 120 keV) was used for the energy and intensity determinations.

4. Experimental Results

Fig. 1 shows the relevant portion of the gamma-ray spectrum. A 5.0 mm Pb absorber was used in order to obtain a sufficiently high counting rate in the energy region of interest without an excessive counting rate of smaller pulses due to low energy gamma rays and x-rays. Energies and relative intensities of the observed gamma-rays are listed in Table 1. The well known energies of the prominent Np^{239} lines[1,2]) served to calibrate the energy scale below 300 keV; above this energy the known lines of Lu^{177m} [10]) and annihilation radiation were used.

The intensity determination was made as follows: The intensities of all new lines were determined relative to that of the 434.7 keV line, by fitting a standard peak shape to the lines and using the fitted peak height, then correcting for absorption in the Pb and for the variation of efficiency with energy of the Ge(Li) detector[6]). The intensity of the 434.7-keV gamma ray was related to those of the previously known lower energy gammas by observing a part of the spectrum that included both the 434.7 and 334.3 keV gammas. It was not desirable to include the entire spectrum (100-600 keV) on one gain setting because the insufficient number of channels per keV would have caused the peak shape analysis to be less accurate.

Although the chemical purification was considered to be thorough, verification that the new gamma-rays belong to Np^{239} decay was obtained by noting that all peaks decayed with the same half-life. The value 2.3 ± 0.2 days was obtained, which is in agreement within our errors with the accepted value.

5. Interpretation of New Levels

When the energy values of the new gamma-rays are summed with the known energy levels of Pu^{239} it is found that they define three and possibly four new levels plus one previously known level. These levels are shown in fig. 2.

Although the amount of information obtained was quite limited because of the weakness of the new radiations (see Table 1), it has been possible to reach some definite conclusions regarding the nature of the newly-observed levels in Pu^{239}.

The following evidence indicates that the four new levels belong to a rotational band based on a K = 1/2 level at 469.8 keV: From the energies of the 469.8, 492.2, and 505.3 keV levels one can fit the simple Bohr-Mottelson equation for a K = 1/2 band

$$E_I = \frac{\hbar^2}{2\mathfrak{J}} [I(I + 1) + a (-)^{I+1/2} (I + 1/2)] \qquad (1)$$

with the parameters $\hbar^2/2\mathfrak{J}$ = 5.043 keV and $\underline{a}$ = 0.48. With these parameters the energy of the fourth member of the band (I = 7/2) is calculated to be 557 keV. If one assigns the observed 392.4 and ~498.7 keV gammas as the transitions between this I = 7/2 level and the I = 9/2 and I = 5/2 members of the ground state band respectively, then the experimental energy value of the I = 7/2 level is defined as 556.2 keV. The agreement between experimental and theoretical values is as close as can be expected here since only the simplest two-term equation for a K = 1/2 band was used to obtain the theoretical value.

An independent check on the K- value of the upper band can be made by comparing the experimental and theoretical relative intensities of photons connecting this band with the K = 1/2 (ground) band. In the usual case, where $L < K_i + K_f$, the theoretical reduced transition probability ratios are given simply by geometric factors[11]:

$$\frac{B(L,I_i \rightarrow I_f)}{B(L,I_i \rightarrow I_{f'})} = \frac{\langle I_i L K_i K_f - K_i | I_i L I_f K_f \rangle^2}{\langle I_i L K_i K_f - K_i | I_i L I_{f'} K_f \rangle^2} \qquad (2)$$

However, in the special case (such as this) where $L \geq K_i + K_f$ the ratios of reduced transition probabilities involve both intrinsic and geometric factors, and are given by:

$$\frac{B(L,I_i \rightarrow I_f)}{B(L,I_i \rightarrow I_{f'})} =$$

$$\left[\frac{\langle I_i L K_i K_f - K_i | I_i L I_f K_f \rangle + b(-)^{I_f + K_f} \langle I_i L K_f, -K_f - K_i | I_i L I_f - K_f \rangle}{\langle I_i L K_i K_f - K_i | I_k L I_{f'} K_f \rangle + b(-)^{I_f + K_f} \langle I_i L K_i, -K_f - K_i | I_i L I_{f'} - K_f \rangle}\right]^2 \qquad (3)$$

where b is a parameter that depends on the intrinsic wave functions[11]). The calculation of b from theory involves considerable uncertainty, so we have treated it as an arbitrary parameter.

In Table 2 the experimental branching ratios from each level of the new band to levels of the ground band are compared with the theoretical ratios for $K_i = K_f = 1/2$. The theoretical ratios, for dipole transitions, were computed for several values of b and it was found that satisfactory agreement between theory and experiment is obtained with use of the value $b = 0 \pm 0.1$. This was the only situation that was found to produce agreement; the experimental intensities could not be accounted for by a choice of b outside the indicated range, nor by the assumption of quadrupole radiation, nor by the use of any value of K_i other than 1/2.

The failure to observe a transition between the 7/2 level of the upper band and the 7/2 level of the ground band can be understood from the information in Table 2: the predicted theoretical intensity of this transition is only 5% of the intensity of the $7/2 \rightarrow 9/2$ transition (392.4 keV), and this

would have been too weak to observe in our experiments.

It is of interest to know the parity of the upper K = 1/2 band. Since that of the ground state K = 1/2 band is <u>even</u>, a knowledge of whether the de-exciting radiations from the upper band are electric or magnetic dipole would define its parity. It is possible in some cases for even a crude determination of the internal conversion coefficients to aid in distinguishing between electric and magnetic dipole transitions. A measurement was attempted, with use of a simple "conversion-coefficient spectrometer" utilizing both Ge(Li) and Si(Li) detectors[6]), but because of interference from the beta spectrum in the region of these very weak lines we were unable to set a sufficiently low limit on the conversion coefficients to allow the choice to be made. In this case, however, one can make use of the results of the scan of the Np^{239} electron spectrum made at high resolution by Ewan, Graham, Geiger, and MacKenzie[2]) to estimate upper limits for electron intensities in the region of interest. From their published intensities we conclude that Ewan et al. should have been able to observe the K- line of the 434.7 keV transition (3/2 → 5/2) if its conversion coefficient is greater than 0.14. The tables of Sliv and Band[12]) list the following theoretical K-conversion coefficients: E1, 0.019; M1, 0.29. We think it likely, therefore, that the 434.7 keV transition is an electric dipole and that the parity of the upper K = 1/2 band is <u>odd</u>. This conclusion would be invalidated if the 434.7 keV transition were an M1-E2 mixture with $\geq$ 40% E2 admixture, but according to the branching ratio information given in Table 2, so large an amount of E2 admixture is unlikely.

On the basis of the present information, we propose that the major component of the newly observed K = 1/2 level at 469.8 keV is the intrinsic particle state labelled 1/2- [501] in the Nilsson scheme[13]). Although this state has not yet been reported in studies of nuclei in the heavy-element

region, recent calculations by Poggenburg, Mang, and Rasmussen[14]) indicate that it should lie at ~0.7 MeV in Pu^{239}. Support for this assignment is given by the fact that the theoretical decoupling parameter (a, in eq. (1)) for state 1/2- [501] is calculated (η = 3) to be + 0.9,[15]) which is reasonably close to the observed value + 0.48. The only other particle states in this energy region are expected to be the even-parity state 1/2+ [620] with a_{theor} = + 1.5 and the odd-parity state 1/2- [761] with a_{theor} = - 5.3. Neither of these alternative assignments is consistent with the experimental data. We cannot however exclude the possibility that the observed state is an octupole vibrational excitation rather than an intrinsic particle excitation, or that it is of mixed vibrational-intrinsic character.

The new data are summarized in fig. 3, which also includes the previously known levels of Pu^{239} grouped into the respective rotational bands.

6. Beta Decay Log Ft Values

In order to infer beta decay log ft values from the measured photon intensities, it is necessary to make some assumptions regarding unseen transitions. In the case of the upper K = 1/2 band we have observed only the inter-band transitions corresponding to decay on the ground K = 1/2 band, and we cannot set meaningful limits on the intensities of the low-energy intra-band transitions, which would be masked by the many known lines of high intensity in the low-energy region. However it seems reasonable to assume that the intra-band transitions are weak in comparison with the observed inter-band transitions; were this not so, the intensities of the transitions de-exciting the base state of the band (469.8 and 461.9 keV transitions) would be substantially greater than those of the other inter-band transitions, contrary to our observations. With the above assumption, and the assumption of negligible

internal conversion, log ft values for beta decay to the 3/2, 5/2 and 7/2 levels were calculated and are given in Table 3. (This was done by normalizing our gamma-ray intensity data to the electron-gamma intensity scale given in Table 3 of Ewan et al.[2]) and then utilizing the previously known beta branching ratios to place the intensity values on an absolute basis.)

The intensities of transitions from the base state (469.8 and 461.9 keV transitions) are very low and could possibly be accounted for by intra-band feeding, so it is not justified to make log ft calculations with the assumption that their intensity results from primary beta decay. In Table 3 the log ft value for beta decay to the 469.8 keV (I = 1/2) level is therefore given as a lower limit.

The ground state of Np^{239} is considered to be made up principally of the 5/2 + [642] Nilsson orbital, and we have characterized the new K = 1/2 band as 1/2 - [501]. If these two states were pure in K, beta transitions with $\Delta L < 2$ would be forbidden by the K- selection rule ($\Delta L \geq K$). The observed beta transitions would then all be of the so-called first-forbidden "unique" type, and the branching ratios would be given by ratios of squares of vector-addition coefficients[11]). On this basis, it would be predicted theoretically that the spin 1/2 (ground) member of the band would receive about one-third of the total beta decay to the band, whereas in fact the observed relative intensity of this beta group is less than 10%. One concludes that the states are, not unexpectedly, mixed in K-values.

The Nilsson orbital 3/2 + [651] is expected as an admixture in the Np^{239} ground state. Beta decay from this configuration to the 1/2 - [501] state of Pu^{239} could proceed by $\Delta L = 1$ (yes) transitions, but these transitions are expected to be seriously hindered by the asymptotic selection rules (here, $\Delta n_z = 5$). Thus although the beta decays to the upper K = 1/2 band appear to take place via small admixtures in the initial or final state wave functions,

it is not surprising that the transition rates are slow (log ft 9-10) and that the transition intensity ratios cannot be correlated simply with theory for pure K- states.

7. Acknowledgments

Our thanks are due Professor J. O. Rasmussen for several helpful discussions. We acknowledge also the assistance of J. A. Harris in the preparation of sources.

Table 1

Np^{239} transition energies and photon intensities

Transition Energy (keV)		Photon Intensity[c]	
This work[a]	Ewan et al.[b]	This work	Ewan et al.[b]
	44.65		
	49.41		
	57.26		
(62)	61.46		1.5
	67.87		
	88.06		
	106.14		13.4
	106.47		
	166.39		
(182.0)	181.71	0.045	
(209.8)	209.76	2.0	2.4
	226.42		
(228.2)	228.20	6.9	7.46
(254.2)	254.41	0.07	0.07
	272.87		0.052
(277.4)	277.62	8.28 ≡	8.28
(285.2)	285.47	0.48	0.37
(315.5)	315.91	0.88	0.88
(334.1)	334.33	1.21	1.17
392.4[d]		0.00091	
429.5		0.0022	
434.7		0.0072	
447.6		0.00015	
454.2		0.00047	
461.9		0.00091	
469.8		0.00062	
484.3		0.00060	
492.3		0.0035	
497.8		0.0018	
~498.7		---	
504.2		0.00045	

[a]) Energy values in parentheses were measured for identification only, and are not precise.

[b])Reference 2.

[c])Arbitrary intensity units, normalized at the value given by Ewan et al. for the 277.6 keV photon. Approximately, 1 intensity unit = 0.017 photons/β decay.

[d])Our estimated errors for the energies of the new photons range from ± 0.2 keV for the strongest lines to ± 0.5 keV for the weakest lines.

Table 2
Photon branching ratios from levels of the 469.8 keV band to the ground state band ($K_i = 1/2$; $K_f = 1/2$)

I_i	I_f	Theoretical relative intensities[a] Dipole Radiation, calc. with $\underline{b}$ = : +5	+1	+0.2	0	-0.2	-1	-5	±∞	Quadrupole radiation calc. with $\underline{b} = 0$	Experimental relative intensities
1/2	1/2	1.0	0.03	0.22	0.53	1.2	94	6.1	2.1	---	0.7
	3/2	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0
	5/2	---	---	---	---	---	---	---	---	0.85	(<0.15)[b]
3/2	1/2	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0
	3/2	1.6	0.19	0.025	0.18	0.55	30	6.8	2.8	0.93	0.17
	5/2	0.39	0.033	0.73	1.3	2.2	44	4.5	1.3	0.23	2.0
	7/2	---	---	---	---	---	---	---	---	1.1	(<0.025)
5/2	1/2	---	---	---	---	---	---	---	---	3.8	(≤0.05)
	3/2	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0
	5/2	3.8	8.7	0.003	0.068	0.17	0.64	1.6	2.4	2.4	0.083
	7/2	2.5	33	1.5	0.87	0.49	0.028	0.24	0.85	0.32	1.2
	9/2	---	---	---	---	---	---	---	---	1.3	---
7/2	3/2	---	---	---	---	---	---	---	---	64	---
	5/2	6.0	52	2.7	1.6	0.92	0.046	0.50	1.6	4.4	~1.1
	7/2	7.5	27	0.32	0.053	0	0.17	0.86	3.3	30	(<0.1)
	9/2	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0
	11/2	---	---	---	---	---	---	---	---	14	---

[a]The E^{2L+1} energy dependence has been removed.

[b]The intensities in parentheses are upper limits. These lines were not observed.

Table 3

Log ft values of Np^{239} beta decays to members of K = 1/2- band

Level Energy (keV)	% β decay	Log ft
556.2	$\sim 3 \times 10^{-3}$	~9.7
505.3	7.1×10^{-3}	9.7
492.6	19.5×10^{-3}	9.4
469.9	$\leq 2.6 \times 10^{-3}$	≥ 10.4

References

1) J. M. Hollander, W. G. Smith, and J. W. Mihelich, Phys. Rev. 102 (1956) 740

2) G. T. Ewan, J. S. Geiger, R. L. Graham, and D. R. MacKenzie, Phys. Rev. 116 (1959) 950

3) W. G. Smith, W. M. Gibson, and J. M. Hollander, Phys. Rev. 105 (1957) 1514

4) F. Asaro, S. G. Thompson, F. S. Stephens and I. Perlman, Bull. Am. Phys. Soc. 8 (1957) 393

5) H. W. Lefevre, E. M. Kinderman, and H. H. Van Tuyl, Phys. Rev. 100 (1955) 1374

6) H. T. Easterday, A. J. Haverfield, and J. M. Hollander, Nucl. Instr. Methods (in press)

7) F. S. Goulding and D. Landis, Proceedings of conference "Instrumentation Techniques in Nuclear Pulse Analysis," Monterey, April 1963, N.A.S.-N.R.C. publication 1184

8) F. S. Goulding, UCRL-11302 (February 1964)

9) F. S. Goulding and W. L. Hansen, UCRL-11261 (February 1964)

10) P. Alexander, F. Boehm, and E. Kankeleit, Phys. Rev. 133 (1964) B284

11) G. Alaga, K. Alder, A. Bohr, and B. R. Mottelson, Dan. Mat. Fys. Medd. 29 (1955) no. 9

12) L. A. Sliv and I. M. Band, Coefficients of Internal Conversion of Gamma Radiation (USSR Academy of Sciences, Moscow-Leningrad, 1956) Part I: K Shell, Part II: L Shell.

13) S. G. Nilsson and B. R. Mottelson, Mat. Fys. Skr. Dan. Vid. Selsk. Vol. 1, No. 8 (1959)

14) J. K. Poggenburg, H. J. Mang, and J. O. Rasmussen, unpublished results (August 1964)

15) J. Valentin, unpublished report (Laboratoire Joliet-Curie de Physique Nucleaire, Orsay)

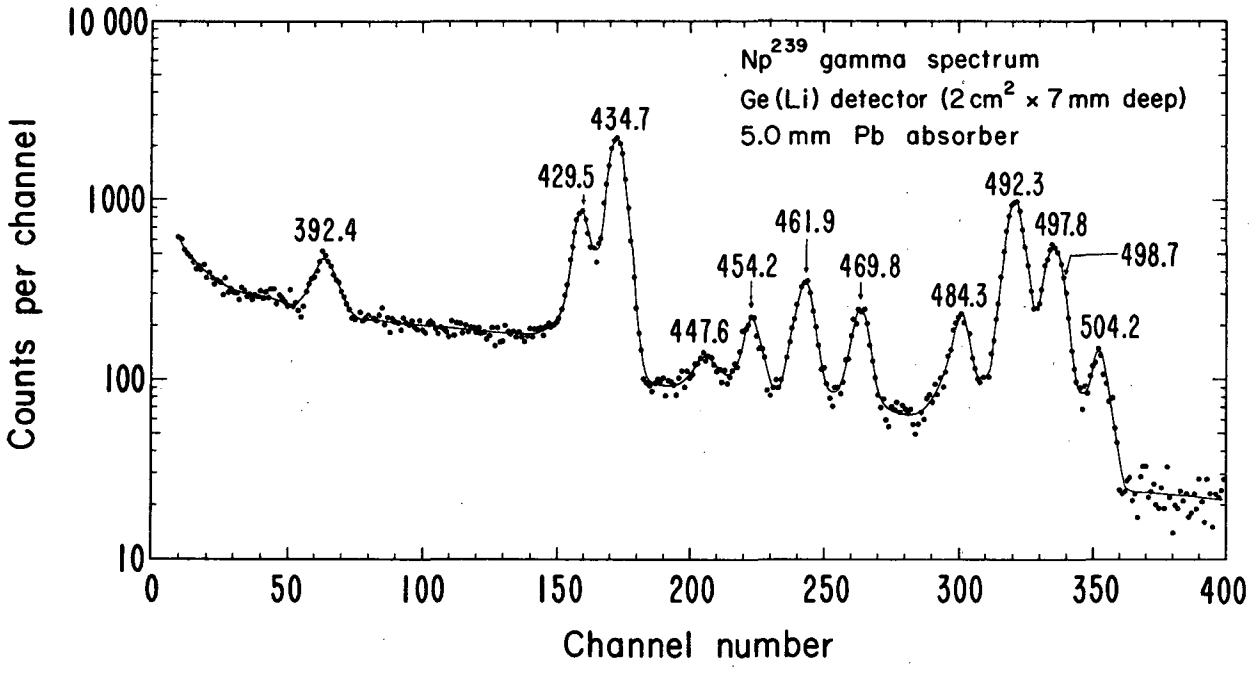
16) J. M. Hollander, Phys. Rev. 105 (1957) 1518

Figure Captions

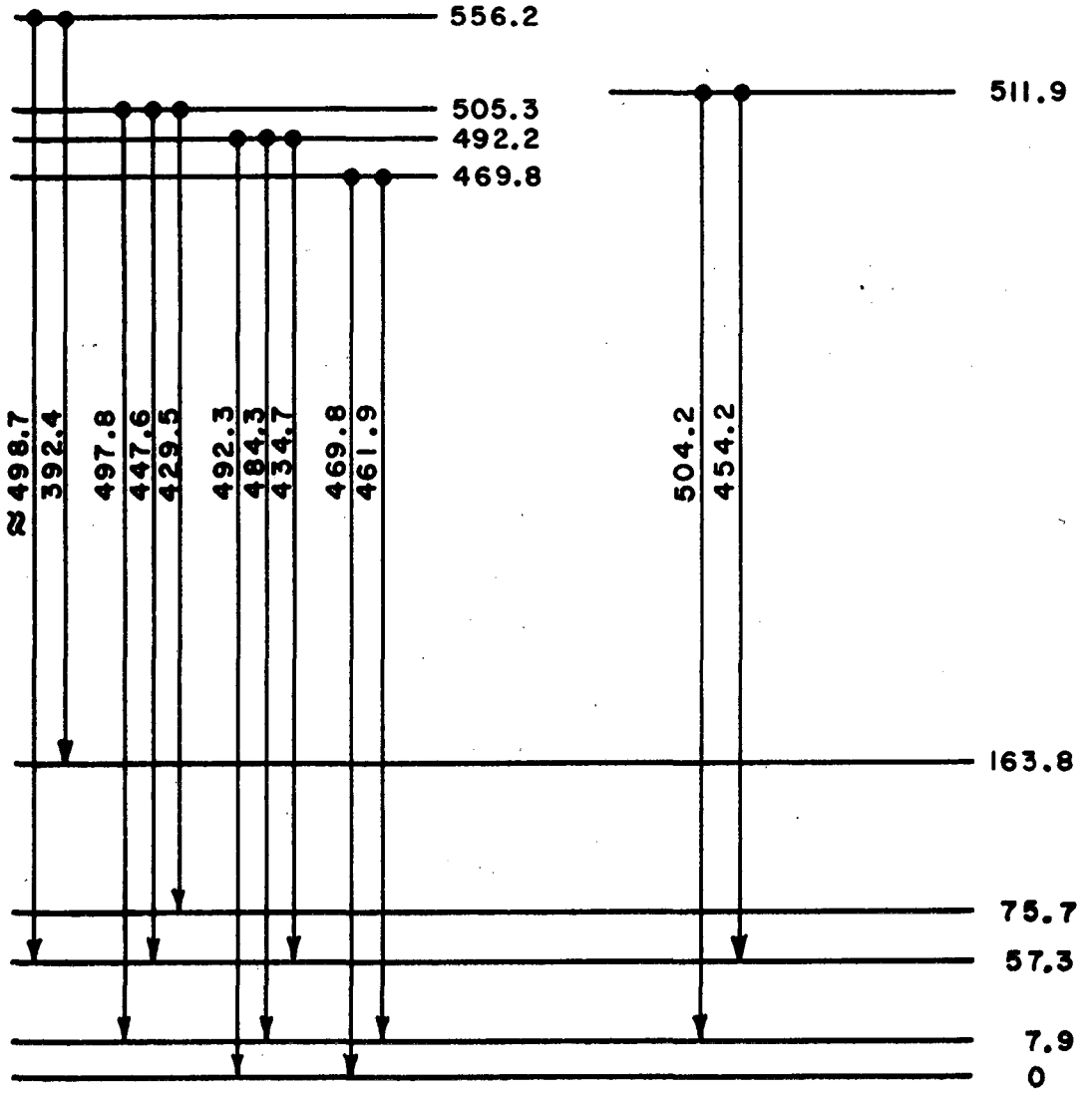
Figure 1. Portion of the gamma ray spectrum that includes the new gamma rays of Np^{239}. The spectrum was observed with a Ge(Li) detector of dimensions 2 cm^2 × 7 mm deep. A 5.0 mm Pb absorber was interposed between source and detector.

Figure 2. Levels of Pu^{239} defined by the new gamma rays observed in this study.

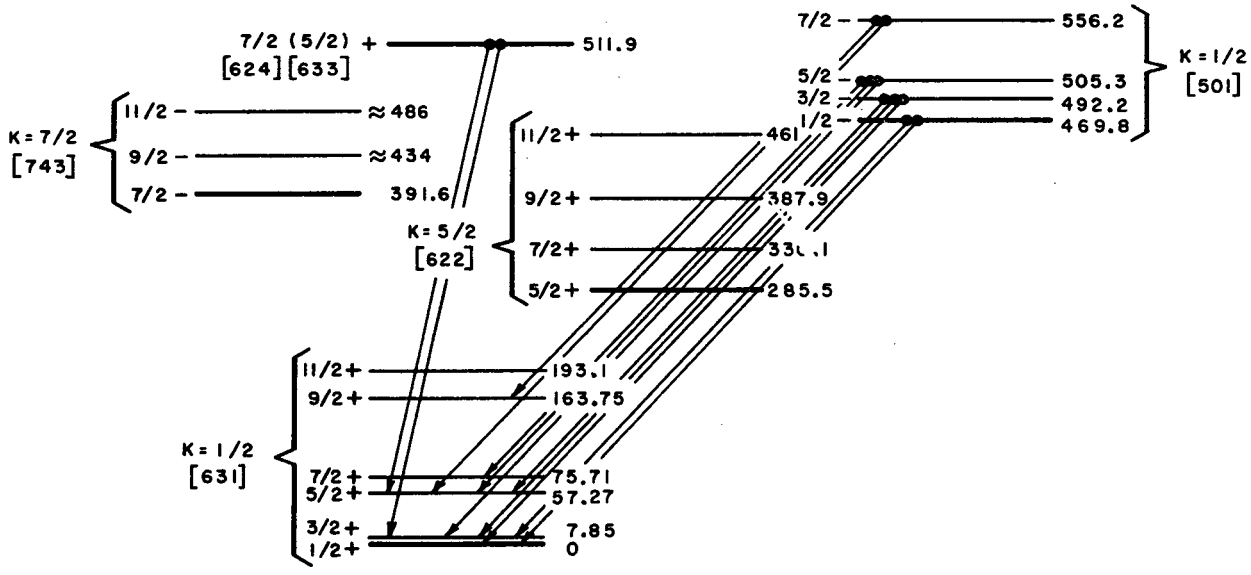
Figure 3. Level scheme of Pu^{239}, showing all known levels grouped into the respective rotational bands. Only the newly-observed transitions are included in this drawing.


Np239 gamma spectrum
Ge(Li) detector (2 cm2 × 7 mm deep)
5.0 mm Pb absorber
392.4
429.5
434.7
447.6
454.2
461.9
469.8
484.3
492.3
497.8
498.7
504.2
Counts per channel
10 000
1000
100
10
0
50
100
150
200
250
300
350
400
Channel number

MUB-4328


556.2
505.3
492.2
469.8
511.9
≈498.7
392.4
497.8
447.6
429.5
492.3
484.3
434.7
469.8
461.9
504.2
454.2
163.8
75.7
57.3
7.9
0

MUB-4326


7/2 (5/2) +
[624] [633]
511.9
K = 7/2
[743]
11/2 −
≈486
9/2 −
≈434
7/2 −
391.6
K = 5/2
[622]
11/2+
461
9/2+
387.9
7/2+
5/2+
285.5
K = 1/2
[631]
11/2+
193.1
9/2+
163.75
7/2+
75.71
5/2+
57.27
3/2+
7.85
1/2+
0
7/2 −
556.2
5/2 −
505.3
3/2
492.2
1/2 −
469.8
K = 1/2
[501]

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