Conversion Electron Spectrum of Np^{239} and Level Scheme of Pu^{239}

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The conversion electron spectrum of Np^{239} has been studied at resolution settings of 0.1% and 0.05% with a 100-cm radius air-cored $\pi\sqrt{2}$ β-ray spectrometer, using a proportional counter as detector. One hundred and forty-three lines were identified corresponding to Auger transitions or the internal conversion of γ rays of energies 44.65, 49.41, 57.26, 61.46, 67.86, 88.06, 106.14, 106.47, 181.7, 209.8, 226.4, 228.2, 254.4, 272.9, 277.6, 285.5, 315.9, and 334.3 kev. The results require the addition of a level at 163.75 kev to the basic level scheme for Pu^{239} proposed by Hollander *et al.* in order to account for three of the weak gamma transitions observed in this work. The relative conversion line intensities are used to determine the transition multipolarities, mixing ratios and expected quantum intensities assuming the K and L shell theoretical conversion coefficients of Sliv. The precisely determined level spacings and the relative transition probabilities are compared in some detail with those expected from an interpretation of the Pu^{239} level scheme in terms of the "Unified Model."

New and more accurate K, L, M, and N electron subshell binding energies for Pu have also been deduced from the experimental data of this work. These values are 20 to 100 ev higher than the values tabulated by Hill, Church, and Mihelich, which were estimated by extrapolation from data at lower Z.

I. INTRODUCTION

PLUTONIUM–239 is in the heavy element region ($A>225$) where nuclei have been found to have pronounced spheroidal deformations and properties characteristic of the "Unified Model."[1] Three rotational bands have been identified in the level scheme of Pu^{239} from studies of the β decay of Np^{239},[2] electron capture decay of Am^{239},[3] and α decay of Cm^{243}.[4,5] The independent-particle states on which these bands are based have been discussed in terms of the Nilsson model by Hollander[6] and Stephens *et al.*[7] The four lowest energy levels in the Pu^{239} level scheme proposed by Hollander *et al.*[2] were interpreted as the members of a ground-state rotational band with $K=1/2$. Weak α-ray groups from Cm^{243} feed what appear to be the fifth and sixth members of this band.[5] The gamma transitions which would be expected to de-excite these two levels have not been identified in earlier investigations. Two other levels have been interpreted as members of a $K=5/2+$ band and another level as a member of a $K=7/2-$ band.

The present paper reports the results of a high-resolution study of the conversion electron spectrum of Np^{239}. This investigation with $\leq 0.1\%$ resolution and high counting rates has revealed a large number of previously unreported conversion lines. Most of these correspond to conversion of known γ transitions in the L and higher subshells. Their intensity ratios were used to determine the multipole mixing ratios of these transitions. Other lines have been assigned to weak γ transitions not previously observed in the β spectrum of Np^{239}. In this way we have identified the fifth member of the ground-state band.

The accurate energies and intensities deduced from these results are compared with the detailed predictions of the Unified Model for the level scheme of Pu^{239}. Of particular interest is the energy of the fifth member of the ground-state rotational band, which has been established by identifying one transition which feeds it and two which de-excite it. This level energy reveals a distinct departure from the spacings predicted by the simple two-parameter formula for a $K=1/2$ band. Even when a correction term for rotational—vibrational interaction is included, there appear to be small but significant discrepancies. The relative experimental $E2$ transition probabilities between members of this band are compared with the theoretical estimates for a $K=1/2$ band and found to agree within experimental error. The observed interband transition probabilities are also compared with the theoretical predictions of the Unified Model.

II. EXPERIMENTAL PROCEDURE

Np^{239} was prepared by irradiating U^{238} (depleted in U^{235}) in the NRX reactor for periods of approximately one day. The procedure used to separate Np^{239} from the inactive uranium and fission products has been described fully in a previous paper.[8] Sources were prepared by subliming the separated Np through a slot 20 mm×2 mm wide onto a backing of 800 $\mu g/cm^2$ Al foil in a vacuum. The initial strength of the source used in the first survey of the conversion electron spectrum was ~1 mC. The surface density of the active material was estimated to be ~30 $\mu g/cm^2$; it was almost invisible. For the experiments at 0.05% resolution, narrower sources were made by subliming through a slot 20 mm ×1 mm wide, and one 0.2-mm wide source was prepared to test the spectrometer at good resolution.

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[1] B. R. Mottelson and S. G. Nilsson, Kgl. Danske Videnskab. Selskab. Mat.-fys. Skr. **1**, No. 8 (1959).

[2] Hollander, Smith, and Mihelich, Phys. Rev. **102**, 740 (1956).

[3] Smith, Gibson, and Hollander, Phys. Rev. **105**, 1514 (1957).

[4] Asaro, Thomson, and Perlman, Phys. Rev. **92**, 694 (1953).

[5] Asaro, Thomson, Stephens, and Perlman, Bull. Am. Phys. Soc. **2**, 393 (1957).

[6] J. M. Hollander, Phys. Rev. **105**, 1518 (1957).

[7] Stephens, Asaro, and Perlman, Phys. Rev. **113**, 212 (1959).

[8] Ewan, Geiger, Graham, and MacKenzie, Can. J. Phys. **37**, 174 (1959).

The conversion electron spectrum was studied in an air-cored $\pi\sqrt{2}$ β-ray spectrometer. A brief description of the instrument has been given previously.[8,9] A more complete description is now in preparation. Most of the experimental results described in this paper were obtained from the first 2-mm wide source. The resolution was set at 0.1% in momentum with a spectrometer transmission of 0.35% of 4π. For the later measurements made with the 1-mm wide source, the resolution was reset to 0.05% with a transmission of ~0.2%. The instrument was calibrated with similar line sources of Th(B+C+C″) and Cs^{137} using the momenta listed by Wapstra *et al.*[10]

The detector used in these experiments was a continuous flow methane proportional counter of conventional design.[11] It has a 1-in. diameter window of 0.9 mg/cm² Mylar plastic film and was coated on one surface with a thin conducting layer of Aquadag colloidal graphite to prevent charging. The transmission of the window as a function of incident electron energy was calculated by interpolation from the transmission curves of Lane and Zaffarano.[12] The curve used for the purpose of this work is shown in Fig. 1. A curve deduced in this manner does not necessarily give the transmission for our particular counter window with high accuracy, but a number of checks were carried out to test its general validity. One test was made by examining the departure from linearity of a Fermi plot of the spectrum of Au^{198} and Au^{199} at low energies. In another test the absorption at 61 kev and 87 kev was measured by observing the reduction in counting rate when an additional 0.9-mg/cm² Mylar film was placed over the window. The low-energy cutoff of ~20 kev is consistent with that expected from the empirical range energy curve for absorbers of ~1 mg/cm². Below 30 kev, where the corrections to be applied are large, intensity measurements obtained using this curve are necessarily approximate.

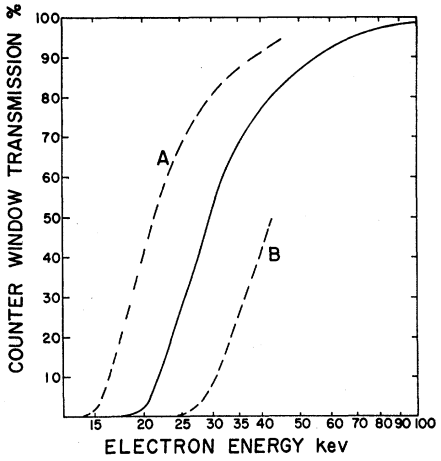

FIG. 1. Solid line shows the counter-window transmission curve used in computing the line intensities listed in Table I. This curve for a 1.1-mg/cm² window was obtained by interpolation from the curves of Lane and Zaffarano.[12] The validity of this curve for our particular window is discussed in the text. For comparison dashed lines *A* and *B* show Lane and Zaffarano's experimental results for a 0.63-mg/cm² window, and for a 1.57-mg/cm² window, respectively.

The focused electrons passed between rectangular edged 0.040-in. thick brass plates which define the counter aperture. The slit was 25 mm×2.5 mm wide for the experiments at 0.1% resolution and 25 mm×1.2 mm wide for those at 0.05% resolution. The slight difference in penetration of the edges of the slit by electrons of widely differing energies may have caused a small systematic error in the relative line intensities quoted below. This effect is believed to have negligible importance, but has yet to be investigated in detail. No correction for it has been applied to the data of this paper.

The background counting rate in the proportional counter was measured periodically, usually every 10 minutes, with the spectrometer current turned off or with the current on and a 1/4-in. Lucite shutter placed in front of the source. The counting rate in both cases was ~40 counts/min. The counting rate with the spectrometer current set to focus electrons above the end point of the β spectrum was also ~40 counts/min. These results suggest that the effect of scattering in the vacuum chamber is small. As a continuous check for possible variations in the counter background contribution the counting rate of a nearby Geiger counter was monitored using a counting rate meter and pen recorder.

The current for the spectrometer coils is derived from a 600-volt dc generator and accurately regulated by a stabilizing system which maintains the voltage drop across a precision standard resistor within ~1 part in 10^5 of the reference potential. In these experiments this potential was derived from a Leeds and Northrup type *K*-2 potentiometer. For all scans it was adjusted manually and the reference voltage was usually advanced in steps of 0.02% to 0.03%. The initial source was strong (~1 mC) and gave high counting rates. Accordingly short counting periods of 15 sec were used. The resulting experimental data were subsequently corrected for background, for decay and when necessary for the 16-μsec dead time of the counting system.

III. TREATMENT OF THE DATA

A vast amount of data was accumulated during the course of these experiments. The procedures used in the analysis of the data are outlined in this section.

The momentum assignments of clearly resolved peaks were deduced from the center of the top of the peaks. This is a more reliable method than deducing a momentum setting by extrapolating the high-energy edge

[9] Graham, Ewan, and Geiger, Bull. Am. Phys. Soc. 4, 64 (1959).
[10] Wapstra, Nijgh, and van Lieshout, *Nuclear Spectroscopy Tables* (North-Holland Publishing Company, Amsterdam, 1959), p. 127.
[11] This counter, Atomic Energy of Canada Limited, type *FB*-2, designed by I. L. Fowler of the Counter Development Section, was adapted by him for use with the $\pi\sqrt{2}$ spectrometer.
[12] R. O. Lane and D. J. Zaffarano, Iowa State Report I.S.C.-439, 1953 (unpublished).

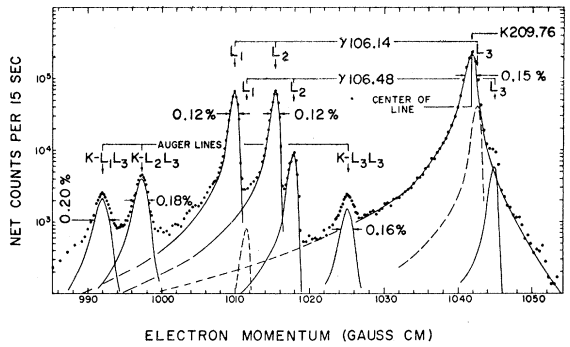

FIG. 2. This demonstrates the manner in which conversion line intensities were derived in this work. After correcting the original data for counter dead-time (16 μsec), background, and decay, the shape of the β continuum was deduced from the line-free portions of the spectrum. The net counts, after subtracting the continuum contribution, are here plotted on a logarithmic scale. This enables closely spaced conversion lines to be decomposed using a standard line shape for L and higher subshells which have natural widths ranging from 7 ev for the L_2 shell to $\sim$30 ev for the M_1 shell. The much greater natural width ($\sim$100 ev) for the K shell gives K conversion lines and K Auger lines a broader shape having a distinct high-energy tail characteristic of the Lorentz factor. The Auger lines, which have three natural-width contributions, appear noticeably broader than the K 209.76 line. The additional width of the K-L_1L_3 peak is due to its complexity.[8] The intensity of the L_3 106.14 (dashed line) is that estimated from multipolarity considerations. The L_1 106.48 peak not resolved here has been identified with 0.06% resolution as shown in the upper right inset of Fig. 4.

to the continuum, since the latter will vary with the natural line width. The peak top center was defined by determining the line width at various heights and extrapolating a smooth line passing up through the half-points to intersect the top of the line as indicated in Fig. 2. This procedure has the advantage of using most of the line data, rather than just the shape of the line near the top, which in some cases was not well defined.

The intensities of the conversion lines could not be determined reliably from peak height, since the line shape was not constant, but depended on the natural width as well as on the energy degradation in the source deposit. Accordingly the areas were determined (divided plot) and corrected for counter window absorption according to Fig. 1. In many cases lines were grouped so closely that simple numerical procedures could not be employed. The usual procedure was to subtract the background and β-continuum contribution and plot the net line counts on semilog paper *vs* momentum as illustrated in Fig. 2. This procedure makes it easy to use the shape of clearly resolved lines in analyzing unresolved groups and makes it possible to estimate the contribution due to the tails of intense lines under nearby weak conversion lines. The K-conversion lines were quite noticeably wider than L and higher subshell lines and also displayed a pronounced high-energy tail. This is due to the much larger natural width, $\sim$100 ev, of a K-shell vacancy. For ease of analysis we have assumed that the L and higher subshell lines had similar shapes although small variations are expected due to differences in their natural widths which range from 7 to 30 ev. This arbitrary procedure may possibly have given rise to small systematic errors, but these have been included in the errors assigned to the intensity values. The K lines were always analyzed with an appropriately broadened shape, since not taking this into account could have led to intensity errors of as much as 30%.

As a check on the intensity deductions procedure outlined above we have found it useful to plot the ratio of peak height to line area against line momentum. The data fall clearly into two groups. A smooth curve rising with momentum can be passed through the points corresponding to L and higher subshells. The K-line points are all lower and lie on a more steeply rising curve, as one would expect from consideration of the greater natural width. These two curves were well defined because of the large number of peak to area ratios deduced in this analysis. Departures from the curves proved useful in revealing arithmetical errors.

It is not practical to present all the data gathered in these experiments in graphical form. A typical portion of the spectrum near 210 kev is displayed in Fig. 3 taken with momentum resolution settings of 0.1%, 0.05%, and 0.013%. The 0.05% resolution was used only when required to resolve closely-spaced conversion lines. The single line shown in the lower part of Fig. 3 demonstrates the capabilities of this instrument for high resolution work. Figure 4 shows the region of the L subshell conversion lines of the two γ rays which de-excite the 9/2+ level at 163.75 kev. In order to establish the $E2$ character of γ 106.47 a search was made for the L_1 conversion line at 0.06%, as shown in the upper right inset. Al-

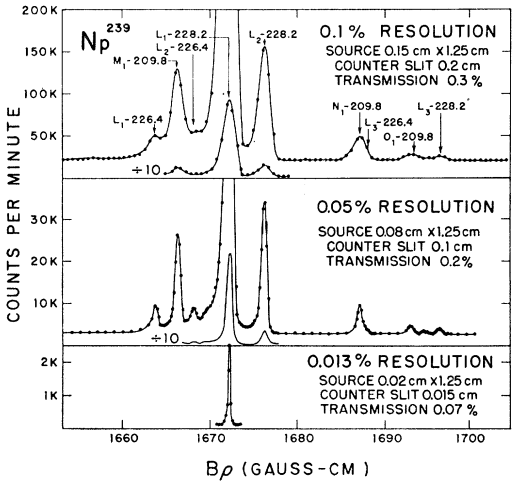

FIG. 3. Conversion electron spectrum of Np^{239} in the region of 210 kev. Most of the results reported in this paper were obtained at 0.10% as in the top part of the figure. When it proved necessary, small regions were rescanned with a resolution setting of 0.05% as shown in the middle part of figure. In this case, the weak L_2-conversion line of the 226.4-kev γ ray is much more clearly resolved from the intense L_1 conversion line of the 228.2-kev γ ray, and hence a more reliable estimate can be made of its intensity. The lower scan shows a test of the spectrometer's capabilities for high-resolution work.

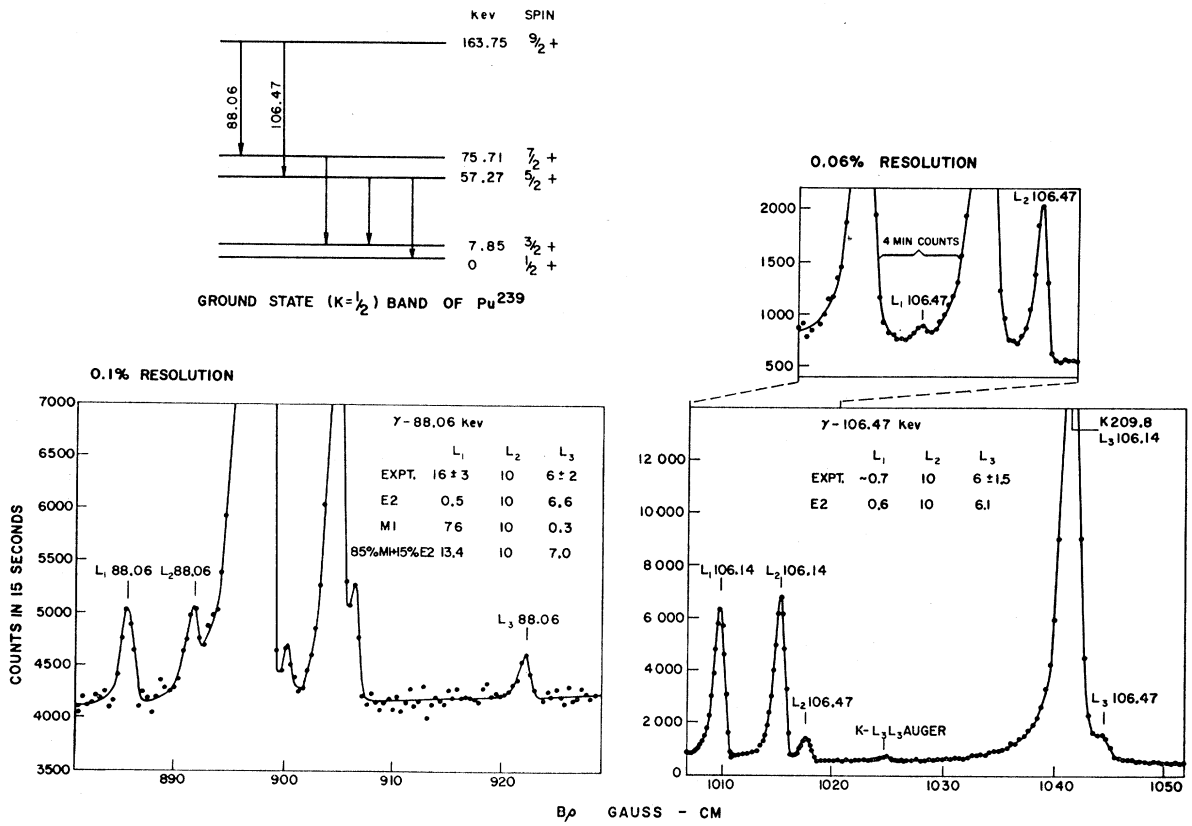

FIG. 4. L conversion lines of the two γ transitions which de-excite the new level at 163.75 kev not previously reported in studies of the β spectrum of Np^{239}.

though the L_1 conversion line is clearly resolved in Fig. 4 the intensity deduced from its area depends critically on the shapes assumed for the two nearby intense lines and is necessarily approximate. However, the $L_1:L_2:L_3$ conversion line ratio clearly establishes the multipolarity of γ 106.47 as predominantly $E2$.

IV. EXPERIMENTAL RESULTS

The conversion line energies and intensities have all been deduced from the data, using the methods described above, and are listed in Table I. Most of these values arise from the initial survey scan at 0.1% resolution. The intensity values for some of the weak lines revealed in later higher resolution scans have been normalized to the original scale by comparison with nearby well-resolved lines. The limit of accuracy for the energy values listed in Table I is estimated to be 1 part in 3000.

The initial assignment of the conversion lines to the various γ transitions was made using the binding energies for $Z=94$ tabulated by Hill, Church, and Mihelich.[13] These values were deduced by extrapolating from the experimental values at lower Z. Hill *et al.*[13] point out that the absolute accuracy of these values is perhaps no better than ~0.1 kev. They did prove accurate enough, however, to make unambiguous assignments to all the lines observed in this work. There is, therefore, no ambiguity in the proposed level scheme for Pu^{239}. The number of conversion lines identified in this work is very much larger than the number of parameters

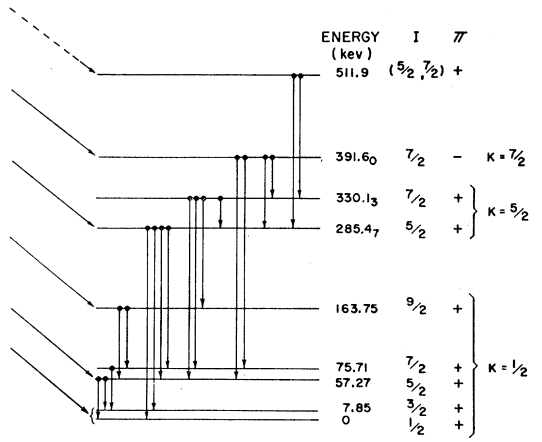

FIG. 5. Proposed level scheme of Pu^{239}. This level scheme is similar to that proposed by Hollander *et al.*[2] for Np^{239} and includes the additional level at 163.75 kev revealed in the present work. The level energies shown here are those deduced from this work and are accurate to ~1 part in 3000.

[13] Hill, Church, and Mihelich, Rev. Sci. Instr. **23**, 523 (1952).

TABLE I. Conversion line spectrum of Np^{239}.

Energy[a] (kev)	Assignment[b]	Intensity[c]
21.48	L_1 44.64	2.4±0.6
22.32	L_2 44.63	0.94±0.2
26.25	L_1 49.41	1.8±0.35
26.54	L_3 44.64	0.71±0.15
27.08	L_2 49.39	2.1±0.4
31.31	L_3 49.41	1.7±0.3
34.11	L_1 57.27	0.35±0.06
34.95	L_2 57.26	7.7±1
38.30	L_1 61.46	0.18±0.04
38.69	M_1 44.67	0.73±0.15
39.17	L_3 57.27	6.5±1
40.05	M_3 44.66	0.18±0.04
43.05	N_1 44.65	0.19±0.04
43.23	L_3 61.33 N_2 44.64	~0.15
43.42	M_1 49.40	0.50±0.1
43.83	M_2 49.42	0.65±0.1
44.28	O 44.61	0.055±0.015
44.54	K 166.39	0.05±0.02
44.78	M_3 49.39 L_1 67.94	0.65±0.1
45.52	L_2 67.83	3.4±0.5
47.83	N_1 49.43	0.18±0.03
48.01	N_2 49.42	0.19±0.03
48.25	N_3 49.40	0.18±0.03
49.09	O 49.42	0.13±0.02
49.32	P 49.39	~0.03
49.74	L_3 67.84	2.4±0.4
51.29	M_1 57.27	0.12±0.03
51.66	M_2 57.25	2.5±0.3
52.66	M_3 57.27	1.7±0.2
53.25	M_4 57.25	0.028±0.007
53.44	M_5 57.26	0.014±0.005
55.48	M_1 61.46	0.048±0.007
55.85	N_2 57.26	0.65±0.08
56.11	N_3 57.26	0.37±0.05
56.34	N_4 57.26	~0.01
56.96	O 57.29	0.30±0.04
57.21	P 57.28	0.06±0.02
59.86	K 181.71 N_1 61.46	0.18±0.03
60.32	N_3 61.47	~0.01
61.14	O 61.47	~0.007
61.88	M_1 67.86	0.04±0.01
62.25	M_2 67.84	0.85±0.1
63.24	M_3 67.85	0.69±0.1
63.86	M_4 67.86	0.014±0.003
64.02	M_5 67.84	0.0065±0.002
64.91	L_1 88.07	0.016±0.002
65.74	L_2 88.05	0.010±0.002
66.41	N_2 67.82	0.24±0.03
66.68	N_3 67.83	0.20±0.03
66.92	N_4 67.84	~0.006
67.54	O 67.87	0.10±0.02
67.78	P 67.85	~0.025
69.98	L_3 88.08	0.006±0.001
75.18	K-L_1L_1	0.087±0.01
75.05	K-L_1L_2	0.167±0.02
76.78	K-L_2L_2	~0.01
80.24	K-L_1L_3	0.052±0.006
81.06	K-L_2L_3	0.097±0.01
82.97	L_1 106.13	0.87±0.09
83.30	L_1 106.46	~0.01
83.81	L_2 106.12	0.93±0.09
84.13	L_2 106.44	0.14±0.02
85.30	K-L_3L_3	0.034±0.004
87.91	K 209.76 L_3 106.11	5.1±0.4
88.38	L_3 106.48	0.086±0.02
92.56 −99.71	K-LM +K-LN	0.18±0.04
100.16	M_1 106.14	0.25±0.03
100.55	M_2 106.14	0.27±0.03
100.91	M_2 106.50	0.045±0.008
101.19	K-L_3N_1	~0.01
101.53	M_3 106.14	0.089±0.015
101.89	M_3 106.50	0.032±0.007
102.18	M_4 106.18	~0.02
102.34	M_5 106.16	~0.01
104.56	N_1 106.16 K 226.41	0.41±0.06
105.06	N_2 106.47	~0.04
106.34	K 228.19	11.9±1
111.58	K-MM	~0.005
132.56	K 254.41	0.105±0.015
143.21	L_1 166.37	0.007±0.001
151.00	K 272.85	0.065±0.01
155.76	K 277.61	9.8±1
158.51	L_1 181.67	0.024±0.004
159.43	L_2 181.74	~0.004
160.42	M_1 166.40	~0.002
163.62	K 285.47	0.040±0.005
175.72	M_1 181.70	0.010±0.003
186.60	L_1 209.76	0.83±0.1
187.43	L_2 209.74	0.11±0.015
191.64	L_3 209.74	0.006±0.002
194.06	K 315.91	0.025±0.004
203.27	L_1 226.43	0.041±0.008
203.79	M_1 209.77	0.21±0.03
204.11	M_2 209.70 L_2 226.42	~0.04
205.04	L_1 228.20	2.15±0.2
205.91	L_2 228.22	0.30±0.04
208.18	N_1 209.78	0.065±0.01
209.41	O 209.74	0.017±0.004
209.77	P 209.84	~0.005
210.13	L_3 228.23	0.012±0.003
212.50	K 334.35	0.029±0.004
220.46	M_1 226.44	0.012±0.003
220.92	M_2 226.48	~0.002
222.22	M_1 228.20	0.54±0.06
222.65	M_2 228.24	0.075±0.01
224.86	N_1 226.46	~0.004
226.59	N_1 228.19	0.16±0.02
227.77	O 228.10	0.045±0.01
228.11	P 228.18	~0.01
231.27	L_1 254.37	0.018±0.003
232.12	L_2 254.43	0.003±0.001
248.55	M_1 254.53	~0.005
249.72	L_1 272.88	0.012±0.002
254.48	L_1 277.64	1.76±0.12
255.30	L_2 277.61	0.23±0.015
259.50	L_3 277.60	0.014±0.003
262.31	L_1 285.47	0.009±0.003
263.15	L_2 285.46	0.030±0.005
266.93	M_1 272.91	~0.003
267.38	L_3 285.48	0.012±0.003
271.64	M_1 277.62	0.41±0.03
272.00	M_2 277.59	0.065±0.01
272.93	M_3 277.54	~0.006
276.05	N_1 277.65	0.12±0.02
277.26	O 277.59	0.036±0.01
277.56	P 277.63	~0.008
279.13	M_1 285.11	~0.002
279.87	M_2 285.46	0.0075±0.001
280.90	M_3 285.51	0.0035±0.001
284.02	N_2 285.43	0.0028±0.001
292.75	L_1 315.91	0.0036±0.001
293.63	L_2 315.94	0.001±0.0004
309.89	M_1 315.87	0.0016±0.001
311.20	L_1 334.31	0.0037±0.001
312.19	L_2 334.35	0.001±0.0004
314.02	N_1 315.62	~0.0007
328.39	M_1 334.37	~0.0015

[a] Based on calibration with F and I lines in ThB spectrum and the K 661 line from Cs^{137} (see Wapstra, Nijgh, and Van Lieshout, *Nuclear Spectroscopy Tables* (North Holland Publishing Company, Amsterdam, 1959). The accuracy limits are about ±1 part in 3000; see text.

[b] The transition energies assigned here were deduced using the electron binding energies determined in this work and listed in Table II.

[c] Relative line areas in arbitrary units after correcting for decay, counter dead-time, variation in line shape, and the assumed counter-window transmission correction shown in Fig. 1. The intensity errors do not allow for an uncertainty in the counter-window correction factor shown in Fig. 1 which was used in calculating the values listed here. For a discussion of this source of error see text.

TABLE II. Binding energies in Pu($Z=94$), in kev.

	K	L_1	L_2	L_3	M_1	M_2	M_3	M_4	M_5	N_1	N_2	N_3	N_4	N_5
Experimental[a]	121.85	23.16	22.31	18.10	5.98	5.59	4.61	4.00	3.82	1.60	1.41	1.15	0.91	
Hill *et al.*[b]	121.75	23.10	22.25	18.05	5.93	5.56	4.56	3.98	3.78	1.56	1.38	1.13	0.86	0.82

[a] A small systematic error may be present due to imperfect cancellation of the earth's field. The estimated absolute accuracy is ±0.05 kev for the K shell and ±0.03 kev for all other shells. The relative accuracy is ±0.03 for the K shell and ±0.020 for all other shells.
[b] Values listed by Hill *et al.* were obtained by extrapolation from lower values of Z.

required to determine the level energies. We make use of this redundancy to deduce, also, the absolute electron binding energies for the Pu atom, $Z=94$. For example the 49.41-kev γ ray is in cascade with the 228.20-kev γ ray, and the crossover 277.62-kev γ ray is also observed. The L_1 binding energy, B.E.(L_1), can be determined from the relation

$$\text{B.E.}(L_1)=E(L_1)_{277}-E(L_1)_{228}-E(L_1)_{49}.$$

There are also several other cascades and cross-overs from which this binding energy may be determined. The mean value of the binding energy B.E.(L_1) is found to be 23.16±0.03 kev. The binding energies of other subshells were deduced from the conversion line energies listed in Table I in a similar manner. The experimental values so obtained are listed in Table II and compared with the values listed by Hill *et al.*[13] The estimated limits of accuracy for our experimentally determined K binding energy is ±0.05 kev and that for the binding energies of the other shells is ±0.03 kev. The fact that the newly determined binding energies are all higher than the tabulated values may indicate that there is a systematic error in the extrapolation procedure used by Hill *et al.* Our experimental values have been used in assigning transition energies listed in Tables I and III.

The errors assigned to the intensities listed in Table I include statistical counting errors, estimates of errors introduced in treating incompletely resolved lines and estimates of errors introduced in determining the contribution from the low-energy tails of conversion lines. These tails could not always be followed to the continuum background because of other conversion lines (see Fig. 2). The errors listed are what we consider to be limits of error due to these causes. It should be noted that we have not included an allowance for the estimated uncertainty in the counter window absorption correction (Fig. 1) used in computing the listed intensities. This method of presenting errors has been chosen since most of our multipolarity assignments depend on the relative intensities of the L subshell conversion lines which are closely spaced in energy. For these deductions the uncertainty due to the variation in the absorption correction is small compared to the absolute uncertainty for all lines in such a group. In comparing the intensity of low-energy lines with high-energy lines, however, an additional error for window absorption should be included which ranges from ~30% at 28 kev through ~10% at 50 kev to less than 2% for energies above 100 kev.

The conversion line intensities and multipolarity assignments of the transitions are summarized in Table III. The multipolarity assignments and mixing ratios have been deduced from the conversion line intensity ratios using the theoretical conversion coefficients of Sliv.[14] In the few cases indicated in Table I, where conversion lines have two assignments, the individual contributions have been deduced, after establishing multipolarities, by using the theoretically predicted subshell ratios. In the case of M and N subshell lines, the ratios used were those observed in these experiments for γ rays of the same multipolarity. The starred quantum intensities listed in the table are those of Ewan *et al.*[15] for the γ rays they observed. For the other transitions the quantum intensities are deduced from the theoretical conversion coefficients. In the case of the $M1$ γ rays this procedure may give rise to small errors as the experimentally measured conversion coefficients[15] do not agree exactly with the theoretical values of Sliv.

V. DISCUSSION OF RESULTS

All the transitions identified in these experiments are accounted for by the level scheme shown in Fig. 4. The high relative accuracy of the transition energies deduced in this work, ~1 part in 5000, together with the total transition intensities provide convincing evidence of the correctness of this level sequence. The absolute accuracy of the level energies is about 1 part in 3000 which includes an allowance for the possibility of error in calibration. This level scheme is basically the same as that proposed by Hollander *et al.*[2] for Np^{239} β decay. Apart from small changes in level energies it differs chiefly in the addition of a 9/2+ level at 163.75 kev. The levels in Pu^{239} can be grouped into a ground-state rotational band with $K=1/2$, a $K=5/2$ rotational band based on the 285.5-kev level, a 7/2− level 391.6 kev ($K=7/2$), and a level at 511.9 kev with spin 5/2 or 7/2. In this section we test how well the Unified Model theory, as presently developed, accounts for the level energy spacings and the relative intensities of competing γ transitions.

The $K=1/2$ ground-state band identified by Hollander consisted of four members seen in their conversion electron study[2] of the Np^{239} β spectrum. The work of Asaro

[14] L. A. Sliv and I. M. Band, Leningrad Physico-Technical Institute Report, 1956 [translation: Report 57 *ICCK*1, issued by Physics Department, University of Illinois, Urbana, Illinois (unpublished)], the L conversion coefficients circulated privately.
[15] Ewan, Knowles, and MacKenzie, Phys. Rev. **108**, 1308 (1957).

TABLE III. Properties of transitions in Pu^{239}.

Transition energy (kev)	Observed line intensities (arbitrary units)[a]													Multipolarity[e]
	K	L_1	L_2	L_3	M_1	M_2	M_3	M_4	M_5	ΣN	ΣO	ΣP	I_γ[b]	I_{tot}	
44.65	...	2.4	0.94	0.71	0.73		0.18			0.19	0.055		0.059	5.3	$0.95M1+0.05E2$
49.41	...	1.8	2.1	1.7	0.50	0.65	0.51			0.55	0.13	~0.03	0.067	8.0	$0.84M1+0.16E2$
57.26	...	0.35	7.7	6.5	0.12	2.5	1.75	0.028	0.014	1.03	0.30	0.06	0.093	20.4	$E2$
61.46	...	0.18		~0.1	0.048					~0.04	~0.007		1.5[d]	2.0	$E1$
67.86	...	0.14	3.4	2.4	0.04	0.85	0.69	0.014	0.0065	0.45	0.10	~0.025	0.083	8.1	$E2$
88.06	...	0.016	0.010	0.006									0.0036	0.036	$0.85M1+0.15E2$
106.14	...	0.87	0.93	0.6	0.25	0.27	0.089	~0.02	~0.01	0.21			13.4*	16.6	$E1$
106.47	...	~0.01	0.14	0.086		0.045	0.032			~0.03			0.025	0.35	$E2$
166.39	0.04	0.007			~0.002								0.008	0.055	$M1$
181.71	0.15	0.024	~0.004		0.010								0.039	0.23	$M1$
209.76	4.5	0.83	0.11	0.006	0.21	~0.03				0.065	0.017	~0.005	2.4*	8.18	$M1$
226.42	0.24	0.041	~0.01		0.012	~0.002				~0.004			0.117	0.42	$M1$
228.20	11.9	2.15	0.30	0.012	0.54	0.075				0.16	0.045	~0.01	7.46*	22.6	$M1$
254.41	0.105	0.018	0.003		~0.005								0.07*	0.20	$M1$
272.87	0.065	0.012			~0.003								0.052*	0.13	$M1$
277.62	9.8	1.76	0.23	0.014	0.41	0.065	~0.006			0.12	0.036	~0.008	8.28*	20.7	$M1$
285.47	0.04	0.009	0.030	0.012		0.0075	0.0035			0.0028			0.37*	0.47	$E2$
315.91	0.025	0.0036	0.001		0.0017					~0.0007			0.88*	0.91	$E1$
334.33	0.029	0.0037	0.001		~0.0015								1.17*	1.2	$E1$

[a] For errors in the conversion line intensities see Table I and discussion in text.
[b] Deduced from theoretical conversion coefficients except the starred values which are experimental values from the work of Ewan, Knowles, and MacKenzie, reference 15.
[c] Deduced mainly from L subshell intensity ratios.
[d] This value may be anomalous; see text.

et al.[5] on the α decay of Cm^{243} showed weak α groups of 5.900 and 5.872 Mev which fed possible fifth and sixth members of this ground-state band. The experiments described in this paper provide evidence for a level at 163.75 kev which is identified as the fifth band member. The assignment of 9/2+ for this level is based on the mixed $M1+E2$ multipolarity of the 88.06-kev transition from it to the 7/2+ level, the pure $E2$ multipolarity of the 106.47-kev transition from it to the 5/2+ level and the absence of transitions to the 3/2+ and 1/2+ levels. It is fed by the 166.39-kev $M1$ transition from the 7/2+ 330.1-kev level and possibly by an $E1$ γ transition of 227.8 kev from the 7/2− level at 391.6 kev. The conversion lines of the latter are expected to be weak and they could not be resolved in our spectra because of the low-energy tails of the intense conversion lines of the 228.2-kev $M1$ γ transition. The 121.71-kev $E2$ transition feeding this level from the 285.47-kev level is expected to be very weak and no conversion lines clearly attributable to it were identified.

Hollander *et al.*[2] fitted the first four levels of the $K=1/2$ band with the simple rotational formula of Bohr and Mottelson,[16]

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

where $\mathcal{I}$ is the effective moment of inertia, I the spin of the level and a the decoupling parameter. The present more accurate energy determinations and the observation of the fifth member of this band show that this description is inadequate. If one deduces the two parameters by fitting to the first three observed level energies, one obtains $\hbar^2/2\mathcal{I}=6.284$ kev and $a=-0.581$. The predicted energies of the fourth and fifth levels are then 75.59 kev and 164.51 kev, respectively. The discrepancies between these predicted values and the observed energies (see Table IV) are well outside any possible experimental error. It is clear that the simple formula (1) is inadequate and that at least one additional parameter is required. One possibility is that rotational-vibrational interaction (RVI) is important here and its inclusion adds an additional term[17] to the level spacing formula:

$$E_I=(\hbar^2/2\mathcal{I})[I(I+1)+a(-1)^{I+\frac{1}{2}}(I+1/2)] -B[I(I+1)+a(-1)^{I+\frac{1}{2}}(I+1/2)]^2. \quad (2)$$

Rather than match the first four level energies, we seek now a "best fit" to all five as shown in Table IV. The discrepancies are still rather larger than the experimental errors but the agreement is much improved. The value of the additional parameter $B=0.0024$ required here is of the same magnitude as the value of $B=0.0034$ deduced by Perlman and Rasmussen[18] for the ground-

[16] A. Bohr and B. R. Mottelson, *β- and γ-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland Publishing Company, Amsterdam, 1955), Chap. XVII.
[17] This form is given by A. K. Kerman, Kgl. Danske Videnskab. Selskab, Mat-fys. Medd. **30**, No. 15 (1956).
[18] I. Perlman and J. O. Rasmussen, *Handbuch der Physik* (Springer-Verlag, Berlin, 1957), Vol. 42, p. 109.

state band in Pu^{238}. There is therefore strong evidence for rotational-vibrational interaction in the odd-N nucleus Pu^{239}. The small but significant energy discrepancies still remaining suggest the presence of another factor which must be taken into account if the theory is to provide an exact description of these level spacings.

The measured transition probabilities within this band can also be compared with the theoretical predictions. The reduced $E2$ transition probability expected for a transition $I_i \rightarrow I_f$ within a rotational band, given by Bohr and Mottelson,[16] is

$$B(E2; i \rightarrow f) = (5/16)e^2Q_0^2(I_i 2K0 | I_i 2I_f K)^2. \quad (3)$$

If we assume that the effective quadrupole moment Q_0 is constant for a given band, it is possible to calculate the ratio of the $E2$ transition probabilities from one level to other levels in the band using (3). The theoretical ratios for transitions within the ground-state band are compared with the experimental ratios in Table V and are found to be in good agreement. In all cases the $E2$ quantum intensity was calculated from the experimental L subshell intensities assuming Sliv's conversion coefficients.[14] The conversion lines of the 18-kev $(7/2 \rightarrow 5/2)$ transition were not observed in the present experiments since their energies are below the counter-window cutoff of $\sim$20 kev.

The theoretical reduced $M1$ transition probability for a transition within a $K=1/2$ band given by Nilsson[19] is

$$B(M1; I+1 \rightarrow I) = \frac{3}{64\pi}\left(\frac{e\hbar}{2Mc}\right)^2 (g_K - g_R)^2 4K^2 \times \left(\frac{2I+1}{I+1}\right)[1+b_0(-1)^{I-\frac{1}{2}}]^2, \quad (4)$$

where g_K and g_R are the intrinsic and rotational gyromagnetic ratios and b_0 a parameter analogous to the decoupling parameter a in Eq. (1). Assuming that these quantities are all constant within the band, the ratio of the reduced $M1$ transition probabilities for the two transitions $9/2 \rightarrow 7/2$ and $5/2 \rightarrow 3/2$ is, from (4),

$$\frac{B(M1; 9/2 \rightarrow 7/2)}{B(M1; 5/2 \rightarrow 3/2)} = 10/9 = 1.11. \quad (5)$$

TABLE IV. Energy levels in ground state $K=1/2$ band, in kev.

Level	Simple[a] rot. model	With[b] R.V.I.	Experimental (kev)
1/2	0	0	0
3/2	7.85	7.88	7.85±0.02
5/2	57.27	57.37	57.27±0.03
7/2	75.59	75.65	75.71±0.03
9/2	164.51	163.75	163.75±0.07

[a] $\hbar^2/2\mathcal{I}$ =6.284 kev, a =0.581.
[b] $\hbar^2/2\mathcal{I}$ =6.290 kev, a = −0.582, B =0.0024 kev.

[19] S. G. Nilsson, Kgl. Danske Videnskab. Selskab. Mat-fys. Medd. **29**, No. 16 (1955).

TABLE V. Reduced relative $E2$ transition probabilities in $K=1/2$ band.

	Experimental[a]	Theoretical
$B(9/2 \rightarrow 5/2)/B(9/2 \rightarrow 7/2)$	14±3	16.5
$B(7/2 \rightarrow 3/2)/B(7/2 \rightarrow 5/2)$	···	9
$B(5/2 \rightarrow 1/2)/B(5/2 \rightarrow 3/2)$	3.4±0.2	3.5
$B(E2; i \rightarrow f) = (5/16)e^2Q_0^2(I_i 2K0 \mid I_i 2I_f K)^2$		

[a] $E2+M1$ mixing ratios and $E2$ quantum intensities deduced from conversion line measurements assuming theoretical L subshell conversion coefficients of Sliv.

The ratio of the reduced $E2$ transition probabilities for the same transitions is, from (3),

$$\frac{B(E2; 9/2 \rightarrow 7/2)}{B(E2; 5/2 \rightarrow 3/2)} = \frac{(\frac{9}{2}\,2\,\frac{1}{2}\,0 | \frac{9}{2}\,2\,\frac{7}{2}\,\frac{1}{2})^2}{(\frac{5}{2}\,2\,\frac{1}{2}\,0 | \frac{5}{2}\,2\,\frac{3}{2}\,\frac{1}{2})^2} = 0.30. \quad (6)$$

Using the observed $M1/E2$ mixture in the $5/2 \rightarrow 3/2$ 49.41-kev transition shown in Table III together with relations 5 and 6, one calculates that the $9/2 \rightarrow 7/2$ 88.06-kev transition should be 86% $M1$+14% $E2$. This is in good agreement with the value of 85% $M1$+15% $E2$ deduced from experiment (see Table III).

The levels at 285.4 kev (5/2+) and 330.1 kev (7/2+) are assigned as the first two members of a $K=5/2$ band. The interband transitions from the 5/2+ level to the 3/2, 5/2, and 7/2 levels in the ground-state rotational band are almost pure $M1$ in character, having less than 5% $E2$ admixture. Similarly the interband transitions from the 7/2+ member of the $K=5/2$ band to the 5/2, 7/2, and 9/2 levels in the ground-state band are also dominantly $M1$ in character, with less than 25% $E2$ admixture. Interband dipole transitions with $\Delta K>1$ are theoretically forbidden. This is consistent with the relatively long measured half-life of 1.1×10^{-9} sec for the 285.4-kev level measured by Graham and Bell.[20] The reduced $M1$ lifetimes for the transitions from this level are $\sim10^4$ slower than the single-particle estimate. However, $E2$ transitions are not K forbidden between levels differing by $\Delta K=2$. We note that the $E2$ components of the transitions from the 285.4-kev level are slowed down by a factor of $\sim$100 from the single-particle estimate. A possible explanation of this $E2$ hindrance factor is found in the asymptotic selection rules first proposed by Alaga[21] to explain hindered β transitions in spheroidally deformed nuclei. The most probable Nilsson assignment[19,6,7] of the asymptotic quantum numbers (K,π,N,n_z,Λ) for the 145th neutron in a $K=1/2$ state is 1/2+[6 3 1] and in a $K=5/2$ state is 5/2+[6 2 2]. This assumes a prolate deformation, $\delta\simeq0.24$, as suggested by the study of the properties of odd-N nuclei in this region.[7] According to the asymptotic selection rules as recently tabulated by Mottelson and Nilsson,[1] a $\Delta K=2$ $E2$ transition with $\Delta n_z=1$ and $\Delta\Lambda=1$ is forbidden and is usually hindered by a factor of $\sim10^2$.

[20] R. L. Graham and R. E. Bell, Phys. Rev. **83A**, 222 (1951).
[21] G. Alaga, Phys. Rev. **100**, 432 (1955).

TABLE VI. Reduced relative $M1$ transition probabilities from second rotational band to ground-state band.

	Experimental[a]	Theoretical $K=3/2 \rightarrow K=1/2$	Theoretical $K=5/2 \rightarrow K=3/2$
$(5/2 \rightarrow 3/2):(5/2 \rightarrow 5/2):(5/2 \rightarrow 7/2)$	0.6:1:0.4	0.83:1:0.38	2.32:1:0.16
$(7/2 \rightarrow 5/2):(7/2 \rightarrow 7/2):(7/2 \rightarrow 9/2)$	0.6:1:0.4	0.75:1:0.35	1.41:1:0.24

[a] Deduced from Table III assuming transitions to be pure $M1$.

Asymptotic selection rules have also been invoked[2] to account for the relative intensities of the β groups which feed levels in Pu^{239}.

If one assumes naively that violation of the K selection rule should rigorously suppress $M1$ transitions between pure bands differing by $\Delta K=2$, it is necessary to seek another explanation of the $M1$ transitions which are observed experimentally. One then examines whether their presence is attributable to lack of K purity, i.e., either one or both of the $K=1/2$ and $K=5/2$ bands is not pure but has a small admixture of $K=3/2$ in it due to a nearby $K=3/2$ band. The probable Nilsson assignment for this would be $3/2+[631]$ and it might occur at an energy of ~ 1 Mev as discussed below. There is no experimental evidence at present for such a band but it could not be fed by β decay if its energy were above ~ 700 kev. $M1$ transitions would then be K allowed from the $K=3/2$ component of the wave-functions in the $K=5/2$ band to the members of the $K=1/2$ band, or alternatively from the $K=5/2$ band to the $K=3/2$ component of the ground-state band. To see if either of these hypotheses is tenable, we compare the reduced transition probabilities for these two cases with the experimental ratios in Table VI. We see that there is qualitative agreement between experiment and theoretical predictions for the $K=3/2 \rightarrow K=1/2$ assumption but not for $K=5/2 \rightarrow K=3/2$.

The level at 391.6 kev has spin $7/2-$ and has been assigned a value of $K=7/2-$. The Nilsson diagram[19] indicates that it can be interpreted as being an intrinsic particle state with quantum numbers $(7/2-743)$. The half-life of this level has been measured as 1.9×10^{-7} sec by Engelkemeir and Magnussen[22] which is $\sim 10^9$ longer than that expected for single-particle $E1$ de-excitation. The transitions to the ground-state band involve $\Delta K=3$ and so violate the K-selection rules for $E1$ transitions. While the $E1$ transitions to the $K=5/2$ band do not violate the K selection rules ($\Delta K=1$) they require changes $\Delta N=1$, $\Delta n_Z=2$, and $\Delta\Lambda=1$ which violate the asymptotic selection rules and hence are expected to be hindered. The predicted ratio of the reduced $E1$ transition probabilities from a $K=7/2 \rightarrow K=5/2$ band is, according to Bohr and Mottelson,[16]

$$\frac{B(E1;7/2 \rightarrow 5/2)}{B(E1;7/2 \rightarrow 7/2)}=\left[\frac{(\frac{7}{2}\,1\,\frac{7}{2}\,1|\frac{7}{2}\,1\,\frac{5}{2}\,\frac{5}{2})}{(\frac{7}{2}\,1\,\frac{7}{2}\,1|\frac{7}{2}\,1\,\frac{7}{2}\,\frac{5}{2})}\right]^2=3.4. \quad (7)$$

The reduced experimental ratio for γ 106.14 and γ 61.46 quantum intensities listed in Table III is 1.4 ± 0.6 which is in poor agreement. According to the asymptotic selection rules for anomalous $E1$ conversion listed by Nilsson and Rasmussen,[23] both of these hindered transitions would be expected to have anomalous conversion coefficients. However, the quantum intensity listed for γ 61.46 in Table III was deduced assuming Sliv's theoretical coefficients. On the other hand, the quantum intensity listed for γ 106.14 was deduced from the anomalous L_1 experimental conversion coefficient of Ewan *et al.*,[15] which disagrees with Sliv's predictions. If one assumes that the degree of "anomaly" is the same for these two transitions and that Sliv's theoretical coefficients give the relative values correctly, the reduced experimental ratio is then 2.6 ± 0.8, which is in much more satisfactory agreement with Eq. (7).

The level at 511.9 kev can have a spin and parity of $5/2+$ or $7/2+$ since the de-exciting transitions to the $5/2+$ and $7/2+$ members of the $K=5/2+$ band are predominantly $M1$. The ratio of the reduced $M1$ transition probabilities to these levels from a $5/2+$, $K=5/2$ level and a $7/2+$, $K=7/2$ level are predicted to be 2.5 and 3.4, respectively. The experimental reduced ratio of 1.7 ± 0.5 deduced from Table III assuming these are pure $M1$ transitions favors the $5/2+$, $K=5/2$ assignment. However, Stephens *et al.*[7] in their recent survey of odd-mass nuclei in the heavy-element region have chosen a Nilsson assignment $7/2+[624]$ for the 512 level on the basis of systematics. The arguments for their preference are not strong and the assignment $5/2+[633]$ was also considered.[7] On the other hand, since a rather accurate experimental ratio is needed to distinguish clearly between these two alternatives, we are reluctant to make a definite $5/2+$ assignment. It is interesting to note that if the $5/2+[633]$ assignment is correct one would expect from the Nilsson diagram that a $3/2+[631]$ level should also be nearby at perhaps ~ 1 Mev. This would be near enough to the $K=5/2$ band based on the $5/2+[622]$ level at 285.47 kev to give rise to appreciable band mixing and so account for the observed relative intensities of the K-forbidden $M1$ transitions discussed above.

V. CONCLUSIONS

The energy spacings between the five members of the $K=1/2$ ground-state band in Pu^{239} have been determined with high accuracy in this experimental study of

[22] D. Engelkemeir and L. B. Magnussen, Phys. Rev. **99**, 135 (1955).

[23] S. G. Nilsson and J. O. Rasmussen, University of California Radiation Laboratory Report U.C.R.L.-3889, 1957 (unpublished).

the conversion electron spectrum following β decay of Np^{239}. The simple two parameter rotational energy level formula for a $K=1/2$ band accounts only qualitatively for the experimental values. A greatly improved fit is obtained when a third rotation-vibration interaction term is included. The interaction strength required to give the "best fit" to the experimental spacings is comparable to that required in the even-even nucleus Pu^{238}. There is thus good evidence that rotation-vibration interaction is important in Pu^{239}. With the inclusion of this third parameter the largest difference between the "best fit" values and the experimental ones is 0.10 kev. This discrepancy is still considerably larger than the experimental uncertainty and may be indicating the necessity of including one or more additional correcting terms in the theoretical formula. One possibility would be to take account of rotation particle coupling from a postulated $K=3/2$ band which is not observed. The influence of such a band on the energy spacing has not been explored in this paper although its presence may be indicated for other reasons. The good agreement between the observed transition probabilities within the ground-state band and the theoretical predictions, also confirms the validity of a Unified Model description for this group of rotational levels.

Both the $M1$ and the $E2$ components of the transitions from the $K=5/2$ band to the ground-state band are strongly hindered. K selection rules account for the slowness of the $M1$ components but asymptotic selection rules must be invoked to explain the $E2$ hindrance. Since these transitions are predominantly $M1$, a possible explanation for the violation of the K selection rules has been sought by assuming that either the $K=1/2$ or the $K=5/2$ band is not pure but has a small admixture of $K=3/2$ in it. It is seen that a $3/2+[631]$ level at $\lesssim 1$ Mev is not inconsistent with present knowledge and could provide the necessary admixture in the $K=5/2$ band to account for the observed $M1$ intensity ratios.

The spin and parity of the level at 511.9 kev must be either $5/2+$ or $7/2+$. The intensity ratios of deexciting transitions deduced from the experimental work of this paper favor an assignment of $5/2+[633]$ while the systematics arguments presented by Stephens *et al.*[7] point to an assignment of $7/2+[624]$. Our experimental evidence for $5/2+$ is not considered to be accurate enough to completely preclude an assignment of $7/2+$.

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