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Energy Levels in ^{239}Pu Populated by Electron-Capture Decay of 11.9-h ^{239}Am

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γ singles spectra of mass-separated ^{239}Am(11.9 ± 0.1 h) samples have been measured with high-resolution Ge(Li) detectors. Conversion-electron spectra were measured at 0.1% resolution with a magnetic spectrometer and also with a cooled Si(Li) detector. An $M1$ multipolarity was found for the weak 124.42-keV transition from the previously known 511.84-keV state to the $\frac{9}{2}+$ level at 387.42 keV. The 511.84-keV state in ^{239}Pu is therefore designated as the spin-$\frac{7}{2}$ member of the $\frac{7}{2}+(624)$ Nilsson state. New very weak intraband transitions (101.96 keV, $E2$ and 57.3 keV, predominantly $M1$) depopulating the $\frac{9}{2}+$ state at 387.42 keV support its spin assignment. Atomic electron binding energies for the K, N_{1-4}, O_2, and O_3 shells in Pu were obtained; small revisions in best values for Pu electron binding energies are suggested. Logft values for the electron-capture transitions ^{239}Am → ^{239}Pu were derived.

I. INTRODUCTION

The conversion electrons associated with the electron-capture (E.C.) decay of ^{239}Am ($t_{1/2}$ = 11.9 h) were first investigated by Smith, Gibson, and Hollander (SGH)[1] and the γ rays were studied by Glass, Carr, and Gibson (GCG).[2] The E.C. decay of ^{239}Am populates levels in ^{239}Pu which are also populated by the β^- decay of ^{239}Np and α decay of ^{243}Cm. The β^- decay[3,4] of ^{239}Np and α decay[5,6] of ^{243}Cm have already been extensively investigated. From these studies rotational bands built on $\frac{1}{2}+(631)$, $\frac{5}{2}+(622)$, and $\frac{7}{2}-(743)$ intrinsic states have been well established. The intrinsic states are described in terms of $K\pi(Nn_z\Lambda)$ where K is the projection of the spin on the nuclear symmetry axis, π is the parity, and N, n_z, and Λ are the asymptotic quantum numbers.[7] More recently levels have been identified belonging to the $\frac{1}{2}-(501)$ band[8] with band head at 469.8 keV. However, a level at 511.8 keV which is more strongly populated in ^{239}Am decay has not been well characterized. (See Sec. IV for decay scheme.) The present investigation was undertaken with the aim of determining the spin of the 511.8-keV state and the reduced E.C. half-lives to various Nilsson states.[7]

II. SOURCE PREPARATION

The ^{239}Am samples for the present measurements were prepared by irradiating ~60 mg of ^{237}Np with 30-MeV α particles in the Argonne 60-in. cyclotron. The irradiation time varied from 6 to 10 h and the average α-particle beam current density was 30 μA/cm^2. The irradiated Np was dissolved by heating it with HNO_3 and HF. After boiling off the HF, the material was evaporated to dryness in *aqua regia*. The residue was then dissolved in 1 ml of 9 M HCl and the Np was extracted four successive times with equal volumes of 0.4 F Aliquat-336 in xylene. To the aqueous phase, which contained Am, 1 mg of lanthanum carrier was added and precipitated as hydroxide. The precipitate was then dissolved in a minimum amount of HCl and evaporated to dryness. The residue was then dissolved in 3 M NH_4SCN-0.01 M H_2SO_4 and loaded onto a 2-mm × 6-cm column containing Aliquat-336 chloride absorbed on hydrophobic diatomaceous earth.[9,10] A solution of 1 M NH_4SCN-0.01 M H_2SO_4 was then passed through the column, which removed most of the fission products. The Am was then eluted with 0.02 M H_2SO_4 solution and was loaded onto another column containing di-2-ethylhexylphosphoric acid (HDEHP) adsorbed on hydrophobic diatomaceous earth.[11] This column was operated at 60°C. A 0.1 M HCl solution was passed through the column which removed most of the remaining fission products. The Am was then eluted with 0.5 M HCl solution. All sources of ^{239}Am were deposited using the Argonne isotope separator.

III. EXPERIMENTAL DATA

A. γ-Ray Spectroscopy

Several γ-singles spectra of mass-separated ^{239}Am samples were measured with a 1-cm^3 planar Ge(Li) detector and a 25-cm^3 coaxial Ge(Li) detector. The 1-cm^3 Ge(Li) spectrometer had a thin Be window and had a resolution full width at half maximum (FWHM) of 0.7 keV at 100-keV γ-ray energy. Figure 1 shows the γ-ray spectrum measured with the 1-cm^3 Ge(Li) spectrometer. The 124.4-keV γ ray in the spectrum has not been observed in the decay of ^{239}Np and ^{243}Cm.

The energies and intensities of higher-energy γ rays were measured with the 25-cm^3 Ge(Li) detector. A set of absorbers (0.7 g/cm^2 Pb, 1.1 g/cm^2 Cd, 0.7 g/cm^2 Cu, and 0.4 g/cm^2 Al) was used to absorb lower-energy γ rays and K x rays in order to reduce summing-effect interference. The spectrum thus measured is shown in Fig. 2.

The energies and intensities measured from several spectra are summarized in Table I. Intensities are expressed in percent per ^{239}Am E.C. decay. These were obtained by equating the total γ-ray and conversion-electron intensities populating the ground-state band to 90%. A 10% E.C. feed was assigned to the ground-state band, on the

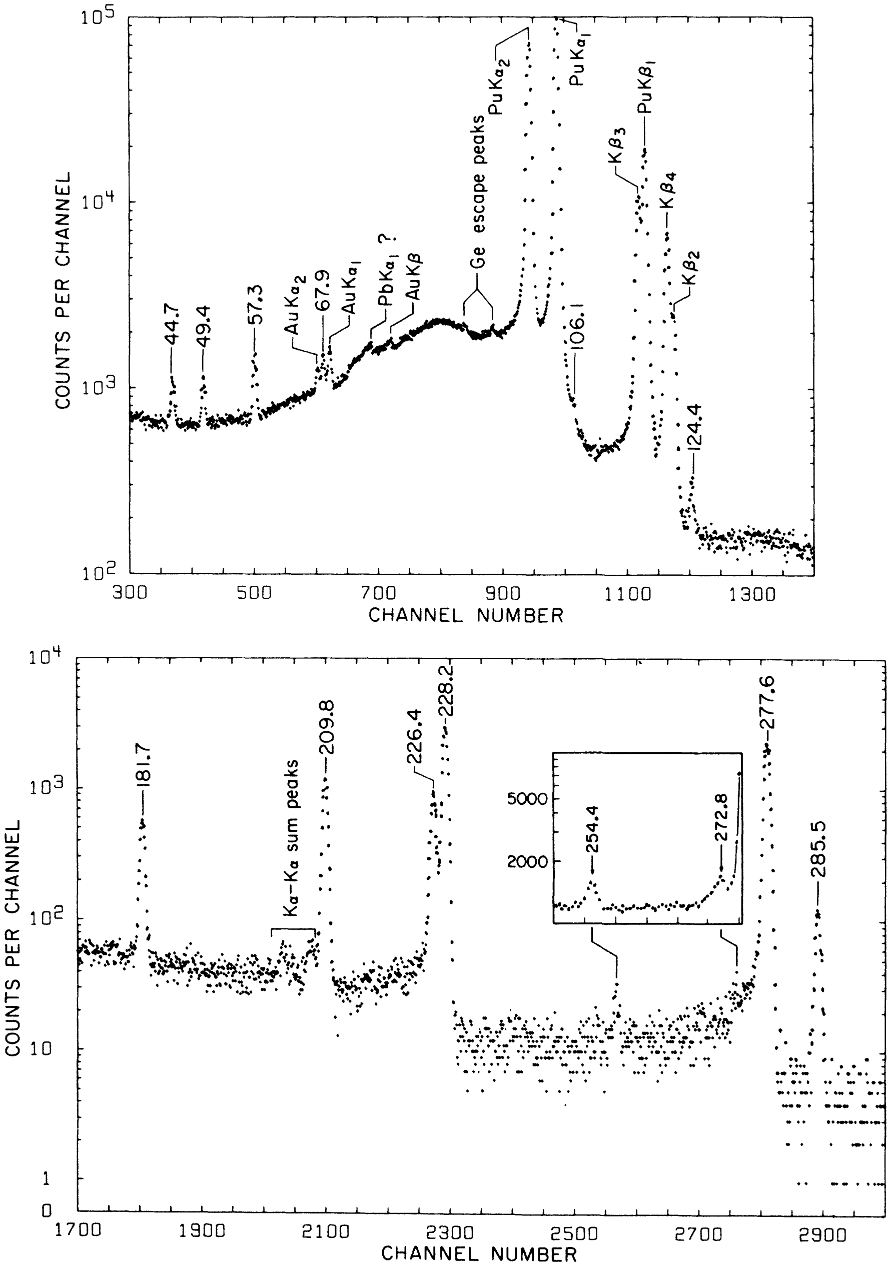

FIG. 1. ^{239}Am γ-ray spectrum measured with a 2-$cm^2 \times$ 0.5-cm Ge(Li) detector. The source was placed 3 cm from the detector endcap, and a 350-mg/cm^2 Al absorber was used to reduce Pu L x rays. Energies are given in keV. Au K x-rays originate from fluorescence of the gold electrode on the detector, and Pb x rays from shielding. Inset shows the region of the 254.4- and 272.8-keV transitions as recorded from the larger 25-cm^3 Ge(Li) spectrometer.

basis of evidence presented in Sec. III C and IV B.

B. Magnetic Electron Spectroscopy

Internal-conversion electrons following the decay of ^{239}Am have been surveyed by SGH[1] using a spectrometer with photographic-plate detector. They noted the absence of the 61.5- and 106.1-keV transitions which are evident in the ^{239}Np decay to ^{239}Pu levels, but found 10 transitions which had previously been seen in the ^{239}Np decay.[3] Our efforts here were to establish some of the weaker decay routes of ^{239}Am. Because of the short half-life we chose to examine only lines critical to the establishment of relative intensity differences in the decay of ^{239}Am and ^{239}Np, and to rely on the relative shell conversion intensities for specific transitions already well determined by Ewan, Geiger, Graham, and MacKenzie (EGGM)[4] in the decay of ^{239}Np.

1. *Instrumentation*

Source material was deposited on a 5-mg/cm^2 Al foil as 200-eV ^{239}Am ions from the decelerated beam of the Argonne electromagnetic mass separator. The beam was decelerated to reduce penetration of the ions into the Al backing. Our experience shows that a source prepared with 200-V ions has very little line broadening above 20-keV electron energy for 0.1% resolution. A mask with a 3-mm-diam hole was used but the focusing was good enough to concentrate the mass in a small spot of 1–2-mm diam. Such a source in the Argonne toroidal-field spectrometers[12] operated in tandem with a 3-mm exit aperture yields a line width (FWHM, momentum) of ~0.11% and a transmission of ~10% of 4π. Source strength was ~4 $\times 10^6$ disintegrations/min; the source was completely invisible. The thickness was <4 μg/cm^2

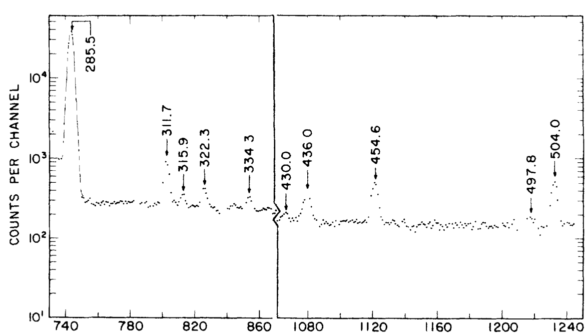

FIG. 2. High-energy portion of ^{239}Am γ-ray spectrum measured with a 25-cm^3 Ge(Li) spectrometer. A set of Pb, Cd, Cu, and Al absorbers was used to absorb the lower-energy γ rays and K x rays. Energies given in keV.

as estimated from the upper limit on background beam current and collection time in the isotope separator.

The detector was a bare cleaved-surface NaI(Tl) crystal scintillation counter whose efficiency was determined by pulse-height analysis at several energies in the range of interest. At 7.3 keV the efficiency is 65%; at 14 keV, 75%; and at 100 keV, 91%. We assign ~10% uncertainty to the counter-efficiency correction in the range of 5–15 keV. A greater uncertainty in low-energy line intensities orginates in energy-loss phenomena in the source material evidenced by a tail on the low-energy side of the line. The area under the tail, the extent of which is the source of the uncertainty, must be included in the line area; this area is proportional to the electron intensity.

We find it useful to plot the ratio of line area to peak height as a function of electron energy to aid in the intensity determination of weaker and not well-resolved lines. Separate curves are generated for K and for L, M, ... conversion lines at energies where the natural width of the K level (115 eV[13] compared with 5–15 eV for L and M lines) becomes a significant fraction of the instrumental resolution. In addition, we find that the

TABLE I. ^{239}Am γ rays and K x rays.

Energy (keV)	Photon intensity (% per decay)	Transition (initial → final level) (keV)
44.7 ± 0.1	$(9 \pm 1) \times 10^{-2}$	330.1 → 285.5
49.4 ± 0.1	0.11 ± 0.01	57.27 → 7.86
57.3 ± 0.1	0.17 ± 0.017	57.27 → 0
67.9 ± 0.1	0.13 ± 0.013	75.70 → 7.86
99.5 ± 0.1	35.0 ± 1.5	Pu $K\alpha_2$
103.8 ± 0.1	53.9 ± 2.3	Pu $K\alpha_1$
106.1 ± 0.2	$(5 \pm 1) \times 10^{-2}$	391.6 → 285.5
117.1 ± 0.1	20.1 ± 1.1	Pu $K\beta_1'$
120.6 ± 0.1	6.9 ± 0.4	Pu $K\beta_2'$
124.4 ± 0.1	0.1 ± 0.01	511.8 → 387.4
181.7 ± 0.1	1.08 ± 0.06	511.8 → 330.1
209.8 ± 0.1	3.5 ± 0.2	285.5 → 75.70
226.4 ± 0.1	3.3 ± 0.2	511.8 → 285.5
228.2 ± 0.1	11.3 ± 0.6	285.5 → 57.27
254.4 ± 0.1	$(8.4 \pm 0.6) \times 10^{-2}$	330.1 → 75.70
272.8 ± 0.1	$(6.4 \pm 0.5) \times 10^{-2}$	330.1 → 57.27
277.6 ± 0.1	15.0 ± 0.7	285.5 → 7.86
285.5 ± 0.1	0.80 ± 0.05	285.5 → 0
311.7 ± 0.2	$(1.7 \pm 0.15) \times 10^{-2}$	387.4 → 75.70
315.9 ± 0.2	$(1.7 \pm 0.15) \times 10^{-2}$	391.6 → 75.70
322.3 ± 0.2	$(2.6 \pm 0.3) \times 10^{-3}$	330.1 → 7.86
334.3 ± 0.2	$(4.2 \pm 0.5) \times 10^{-3}$	391.6 → 57.27
430.0 ± 0.3	$(1.7 \pm 0.3) \times 10^{-3}$	505.7 → 75.70
436.0 ± 0.3	$(8 \pm 1) \times 10^{-3}$	511.8 → 75.70
454.6 ± 0.3	$(1.2 \pm 0.12) \times 10^{-2}$	511.8 → 57.27
497.8 ± 0.3	$(1.5 \pm 0.3) \times 10^{-3}$	505.7 → 7.86
504.0 ± 0.3	$(1.4 \pm 0.14) \times 10^{-2}$	511.8 → 7.86

L Auger lines have a different shape and fall on a still different curve. Line intensity (either from area measurement or by determining area from peak height as described just above) was corrected for decay and counter efficiency.

Energy calibration was derived from the K conversion line of the 122.060-keV transition[14] in ^{57}Fe. Line positions were determined from the medians of the upper quarter of the peaks.

2. *Electron Binding Energies*

Table II gives the binding energies in Pu used in the present work. They represent values selected from smooth curves (modified "Moseley" plots)[15] which are fitted to the experimental data available in the actinides[16–23] from $Z = 92$ to 100 and from this work. For comparison, the values of the Bearden and Burr[16] compilation are tabulated; many of these exhibit deviations of several probable errors from the expected smooth Z dependence.[15] We present our selected values for general utility, primarily because of these large deviations. The Pu binding energies determined by EGGM[4] were not included because they are systematically larger by 30–60 eV due to an instrumental problem the possibility of which was anticipated by their assigning errors of that magnitude.

The present work makes some contribution to the binding energy data in the following way. The energy difference between lines from the same shell of the 49.4- and the 57.3-keV transitions gives the energy of the 7.86-keV transition. From the weighted average of the L_1, L_2, L_3, M_2, and M_3 line-pair differences of these transitions we obtain 7860 ± 3 eV for the energy of the first excited state in ^{239}Pu. This enables us to calculate the N_{1-4} and $O_{2,3}$ binding energies from the measured line energies of the 7.86-keV transition given in Table III. In addition, the K binding energy can be based on the L_1, L_2, and M_1 binding energies and the corresponding conversion lines of the 102- and 124-keV transitions; the sum of these energies yields the 226-keV transition energy. Our result, from the 226-keV K line (see Table II) is in good agreement with the more precise K bind-

TABLE II. Atomic electron binding energies ($Z = 94$) to the Fermi level of solid plutonium oxide, all values in keV ± (eV).

Shell	Selected values [a]	Present work	Bearden and Burr [b]
K	121.803 (7)[c]	121.801 (23)	121.818 (66)
L_1	23.102 (5)		23.097 (2.5)
L_2	22.269 (3)		22.266 (1)
L_3	18.057 (2)		18.056 (1)
M_1	5.930 (3)		5.933 (2)
M_2	5.548 (5)		5.541 (2.5)
M_3	4.563 (5)		4.557 (2)
M_4	3.970 (4)		3.973 (1)
M_5	3.778 (4)		3.778 (1)
N_1	1.564 (4)	1.563 (4)	1.559 (1)
N_2	1.387 (4)	1.387 (4)	1.372 (3)
N_3	1.126 (2)	1.128 (4)	1.115 (2.5)
N_4	0.850 (1)	0.846 (8)	0.849 (1)
N_5	0.801 (1)		0.801 (1)
N_6	0.445 (10)		0.445 (2.5)
N_7	0.433 (10)		0.432 (3)
O_1	0.354 (3)		0.352 (3.5)
O_2	0.285 (6)	0.293 (5)	0.274 (7)
O_3	0.213 (6)	0.221 (4)	0.207 (7)
O_4	0.116 (3)		0.116 (2)
O_5	0.106 (3)		0.105 (1.5)
P_1	0.075 (10)		
P_2	0.040 (10)		
P_3	0.030 (10)		

[a] These values selected on the basis of smooth fits to "modified Moseley plots" of experimental binding energies vs Z in the actinides from $Z = 92$ to 100. The errors indicated are intended to be standard deviations and reflect the uncertainty in the fitting. The data were taken from Refs. 16–23 and the present work.

[b] Reference 16. The error values given here are standard errors adjusted from the probable errors given in Ref. 16.

[c] See Ref. 23 for K binding energy.

TABLE III. Internal-conversion lines in the decay $^{239}Am \rightarrow {}^{239}Pu$ selected for observation with magnetic spectrometer.

Transition keV ± (eV) (initial → final levels)	Shell	Electron energy keV ± (eV)	Transition energy[a] keV ± (eV)	Intensity[b]	Multipolarity: data[c] from which derived
7.860 (3)	ΣM			(33.4)	99.7% $M1$
(7.86 → 0)	N_1	6.297 (2)	7.861 (5)	4.00 ± 0.28	+ (0.30 ± 0.02)% $E2$
	N_2	6.473 (2)	7.860 (5)	2.92 ± 0.25	N_1/N_2
	N_3	6.732 (2)	7.858 (3)	2.86 ± 0.28	N_1/N_3
	N_4	7.014 (7)	7.864 (7)	0.08 ± 0.04	
	N_5	7.063 (11)	7.864 (11)	0.04 ± 0.03	
	O_1			(0.90)	
	O_2	7.567 (3)	7.852 (7)	0.69 ± 0.08	
	O_3	7.639 (2)	7.852 (7)	0.67 ± 0.07	
	$O_{4,5}$			(0.015)	
	$\Sigma P, Q$			(0.40)	
				46.0	
44.663 (5)	L_2	22.394 (3)	44.663 (5)	1.35 ± 0.15	
(330.1 → 285.5)	Other e^-			(10.1)	
	γ			0.09	
				11.5	
49.412 (4)	L_1	26.304 (3)	49.406 (6)	2.20 ± 0.13	80% $M1$
(57.27 → 7.86)	L_2	27.148 (3)	49.417 (5)	3.13 ± 0.19	+ (20 ± 2)% $E2$
	L_3	31.357 (3)	49.414 (4)	2.50 ± 0.14	L_1/L_2
	M_2	43.862 (4)	49.410 (6)	0.88 ± 0.05	L_1/L_3
	M_3	44.850 (5)	49.413 (7)	0.74 ± 0.05	
	Other e^-			(1.81)	
	γ			0.11	
				11.4	
57.273 (4)	L_1	34.174 (4)	57.276 (6)	0.90 ± 0.05[d]	$E2 > 85\%$
(57.27 → 0)	L_2	35.006 (4)	57.275 (5)	10.5 ± 0.5	L_1/L_3
	L_3	39.216 (4)	57.273 (4)	9.1 ± 0.5	
	M_2	51.723 (5)	57.271 (7)	3.19 ± 0.22	
	M_3	52.708 (5)	57.271 (7)	2.82 ± 0.20	
	Other e^-			(2.12)	
	γ			0.16	
				28.8	
67.841 (7)	L_1	44.726 (18)	67.828 (18)	0.15 ± 0.03	$E2 > 75\%$
(75.70 → 7.86)	L_2	45.573 (5)	67.842 (7)	4.5 ± 1.0	L_1/L_2
	Other e^-			(6.0)	
	γ			0.12	
				10.8	
101.965 (13)	L_1			≤0.008	$E2 > 80\%$
(387.4 → 285.5)	L_2	79.701 (13)	101.970 (13)	(0.054)[e]	L_1/L_3
	L_3	83.888 (24)	101.945 (24)	0.034 ± 0.005	
	Other e^-			(0.034)	
	γ			(0.008)	
				0.13	
106.1 (391.6 → 285.5)	L_1			≤0.016	
106.505 (24)	L_2	84.236 (24)	106.505 (24)	0.048 ± 0.005	
(163.8 → 57.27)	Other e^-			(0.069)	
	γ			(0.009)	
				0.126	

TABLE III *(Continued)*

Transition keV ± (eV) (initial → final levels)	Shell	Electron energy keV ± (eV)	Transition energy[a] keV ± (eV)	Intensity[b]	Multipolarity: data[c] from which derived
124.416 (15)	L_1	101.307 (15)	124.409 (15)	0.176 ± 0.013	$M1$
(511.8 → 387.4)	L_2			≤0.03	($E2$ < 7%)
	M_1	118.500 (21)	124.430 (21)	0.044 ± 0.009	L_1/L_2
	Other e^-, mostly K			(1.08)	
	γ			0.10	
				1.4	
181.715 (10)	K	59.912 (7)	181.715 (10)	3.1 ± 0.6	68% $M1$
(511.8 → 330.1)	Other e^-			(0.78)	+ (32 ± 5)% $E2$
	γ			1.08	α_K
				5.0	
223.6	K			≤0.02	
(387.4 → 163.8)	Other e^- + γ			≤0.02	
				≤0.04	
226.383 (12)	K	104.580 (10)	226.383 (12)	5.68 ± 0.27	77% $M1$
(511.8 → 285.5)	Other e^-			(1.6)	+ (23 ± 7)% $E2$
	γ			3.50	α_K
				10.6	
228.184 (12)	K	106.381 (10)	228.184 (12)	21.6 ± 1.0	88% $M1$
(285.5 → 57.27)	Other e^-			(6.0)	+ (12 ± 7)% $E2$
	γ			11.2	α_K
				38.8	
277.604 (16)	K	155.801 (14)	227.604 (16)	16.6 ± 1.0	88% $M1$
(285.5 → 7.86)	Other e^-			(4.5)	+ (12 ± 5)% $E2$
	γ			15.0	α_K
				36.1	
311.7	K			<0.015	
(387.4 → 75.7)					

[a] Calculated with selected binding energies of Table II.

[b] Errors include statistical, scattering-tail, and decay correction uncertainties. Error in counter-efficiency correction (uncertain by ~10% between 5–15 keV; less uncertain at higher energies) not included. Units are % per ^{239}Am decay assuming 10% electron capture to the ground-state band. Values in parentheses are derived quantities either from theoretical internal-conversion ratios or from experimental data of EGGM.[4] Transition intensities are summed.

[c] Theoretical internal-conversion coefficients for N and higher shells from Ref. 27, and for K, L, and M shells from Ref. 28.

[d] Observed line intensity of this L_1 line is composed of ~ equal contributions from the two 57.3-keV transitions. See decay scheme (Fig. 7) and text, Sec. III B 3.

[e] Experimental intensity of 0.086 was associated with unexplained broader line.

ing energy of Nelson, Saunders, and Salem[23] derived from K x-ray measurements.

3. *Results of Electron Spectroscopy*

The energies and intensities of the conversion lines observed in the present study are given in Table III. Support for the level at 387.4 comes from the very low-intensity transitions, 101.9 keV and 124.4 keV. The pertinent regions of the spectrum are shown in Fig. 3. It is not surprising that the 124.4-101.9-keV cascade was not seen by EGGM[4] in the ^{239}Np decay because it constitutes only 8% of the deexcitation of the 511.8-keV level which is populated in only ~1% of the ^{239}Np decays, whereas the 511.8-keV level is populated in 17% of the ^{239}Am decays. Further, the L_3 line of the 101.9-keV transition would be completely masked by the L_2 line of the 106.1-keV transition in the ^{239}Np decay. The line we call L_3-101.9 cannot be the L_2-106.1 because no L_1-106.1 is seen in ^{239}Am, and EGGM[4] have shown that L_1-106.1 and L_2-106.1 have approximately the same intensity. The line L_2-101.9 is clearly wider than normal and has ~60% too large an intensity compared to L_3-101.9 for an $E2$ transition. Because of this unexplained contribution we calculate the intensity of the 101.9-keV transition in ^{239}Am from the L_3 line and the assumption that it is a pure $E2$ as evidenced from the low L_1 intensity.

The weak 124.4-keV transition is almost pure $M1$ according to the L_1/L_2 ratio and L_1 conversion coefficient. In the region just below L_2-124.4 lies any possible K-223.6 (387.4 → 163.8-keV transition). Our limit is ≤0.02% per decay on the K line or ≤0.04% for the 223.6-keV transition.

The 18.429-keV transition between the 75.70- and 57.27-keV levels in the ground-state band has its conversion lines intermixed with L Auger lines. We find lines at the expected positions of the M_1, M_2, M_3, N_1, and N_2 lines of the 18.429-keV transition but not clearly enough resolved to say with certainty what intensity this transition has. Its total transition intensity is not more than 3% per ^{239}Am decay. An intensity of ~3% would be required if our 209.8- and 67.8-keV intensities are correct.

From our selected lines of the well-established transitions the lower-energy levels of ^{239}Pu involved in the ^{239}Am decay can be determined within a few eV and the upper levels to ~25 eV. In the cases where redundant paths can be used to build the level energy, the agreement is consistent with the assigned errors. For example, at the 285.5-keV level, two paths, 277.6 + 7.86 and 228.2 + 57.3, yield 285.462 ± 0.029 and 285.455 ± 0.025 keV, respectively. Also the 511.8-keV level can be derived from either the 330-keV level plus the 181.7-keV transition or the 387.4-keV level plus the 124.4-keV transition; these yield 511.836 ± 0.025 and 511.839 ± 0.030 keV, respectively. We agree with EGGM[4] (within quoted errors) on all the ^{239}Pu

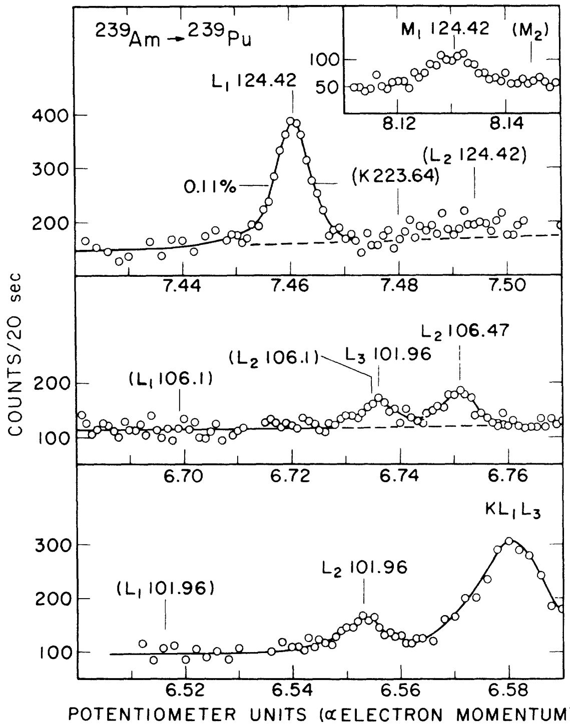

FIG. 3. Internal-conversion lines from the low-intensity 101.96- and 124.42-keV transitions in the decay of ^{239}Am → ^{239}Pu. Expected positions for various lines are indicated by vertical marks. Parentheses indicate lines for which only an upper limit on intensity was obtained. The continuum is the scattering tail of the very intense K lines of the 226.4-, 228.2-, and 277.6-keV transitions at 7.591-, 7.662-, and 9.473-potentiometer units, respectively. Note that the line labeled L_3-101.96 has little contribution from L_2-106.1 because it is known from the decay of ^{239}Np → ^{239}Pu that L_1- and L_2-106.1 have approximately equal intensity.

level energies fed by β decay except, of course, that we saw no electron lines from transitions involving the 391.6-keV level which is populated very weakly in ^{239}Am decay.

From our level energies we predict the energy

TABLE IV. Relative (to L_3 lines = 1 unit) conversion-line intensities for the 49.4- and 57.3-keV transitions following the decay of ^{239}Am and ^{239}Np to ^{239}Pu.

	57.3 keV			49.4 keV	
Shell	^{239}Am Present work	^{239}Np EGGM[a]	Pure $E2$[b]	^{239}Am Present work	^{239}Np EGGM[a]
L_1	0.099 ± 0.008	0.054 ± 0.012	0.048	0.92 ± 0.07	1.06 ± 0.25
L_2	1.16 ± 0.09	1.18 ± 0.23	1.26	1.25 ± 0.10	1.24 ± 0.30
L_3	1	1	1	1	1
M_2	0.35 ± 0.03	0.38 ± 0.07	0.34	0.35 ± 0.03	0.38 ± 0.09
M_3	0.31 ± 0.03	0.27 ± 0.05	0.28	0.29 ± 0.03	0.30 ± 0.08

[a] Reference 4.
[b] Reference 28.

of the intraband transition from the 387.4- to the 330.1-keV level to be 57.302 ± 0.030 keV (we call this the upper 57-keV transition). Conversion lines from this transition would be within a linewidth (~75 eV) of the same lines of the strong 57.273-keV transition. We note first that the upper 57 keV will probably have a large $M1$ contribution as opposed to the pure $E2$ character of the lower 57-keV transition, and second that in the ^{239}Np β decay the relative intensity of the upper 57-keV transition would be 10 times smaller than in ^{239}Am because of the smaller population of the 387.4-keV level following the β^- decay. Table IV shows the line intensities of the 49.4- and 57.3-keV transitions for the ^{239}Am decay from our work and for the ^{239}Np decay from EGGM. It is clear that the 57.3-keV L_1 line is more intense relative to the $L_{2,3}$ and $M_{2,3}$ lines in the ^{239}Am decay. The L_1/L_3 ratio of the 57.3-keV transition is a factor of 2 larger in ^{239}Am than in ^{239}Np, whereas all other line ratios agree to better than 15%, e.g., the 49.41-keV line ratios given in Table IV. Furthermore, the theoretical ratios for a pure $E2$ transition agree with ^{239}Np data rather than the ^{239}Am data. Our interpretation is that this is evidence for the upper 57-keV transition, which has a large $M1$ component. If it were pure $M1$, its intensity, calculated by assigning the 50% excess L_1 intensity to it and by using theoretical conversion coefficients, is 0.7% per decay. This is a lower limit on its intensity, since a small $E2$ admixture can contribute significantly to the upper 57.3-keV components of the L_2 and L_3 lines with little contribution to L_1 and little fractional reduction in the lower 57.3-keV intensity. Figure 4 shows the L-subshell lines from the 57.3- and 49.4-keV transitions. The L_1-57.3 line shows no obvious broadening. Therefore, since it is composed of two approximately equal intensity components the energy of the upper 57-keV transition is within[24] ~10 eV of the lower 57-keV transition, which is consistent with the prediction from the level energies.

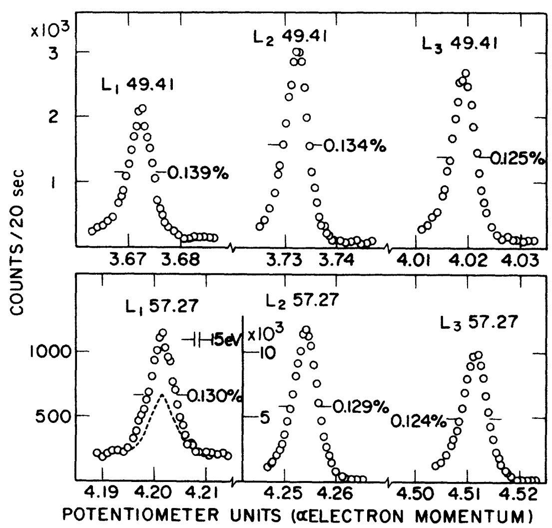

FIG. 4. The L-subshell conversion lines of the 57.27- and 49.41-keV transitions in the ^{239}Am → ^{239}Pu decay. The line L_1-57.26 is a factor of ~2 larger than that expected for a pure $E2$ transition (shown by dashed curve). In the ^{239}Np → ^{239}Pu decay this same transition shows an L_1 intensity expected for a pure $E2$ transition. (See Table IV.) This is interpreted as evidence for another 57.3-keV transition of predominantly $M1$ multipolarity. This second 57.3-keV transition is indeed expected from the level scheme of ^{239}Pu. Note little if any broadening due to the doublet at this resolution. The linewidth increase $L_3 \to L_2 \to L_1$ in both the 57.26- and 49.41-keV cases is due to the progressively larger natural widths of the L_2 and L_1 levels.

Figure 5 shows the region of the $N, O, \ldots$ conversion lines of the 7.86-keV transition. A very similar spectrum is displayed by Ewan and Graham[25] for this transition populated from the Np^{239} decay. Bhalla[26] calculated the N-subshell ratios for this particular transition and concluded that the $E2$ mixing was (0.28 ± 0.07)%. Dragoun, Pauli, and Schmutzler[27] have used the EGGM[4, 25] data with their calculated internal conversion coefficients to derive the $E2$ mixing. Our data give the

TABLE V. Total intensity of the 7.86-keV transition based on theoretical internal-conversion coefficients normalized to the average of the experimental intensities in the $N_{1,2,3}$ subshells. Intensities in percent per ^{239}Am decay assuming 10% electron capture to the ground-state band.

Shell	Theory 0.997 $M1$ + 0.003 $E2$	Experiment
M_1	13.7	
M_2	9.9	
M_3	9.5	
M_4	0.17	
M_5	0.11	
N_1	4.02	4.00 ± 0.28
N_2	2.96	2.92 ± 0.25
N_3	2.81	2.86 ± 0.28
N_4	0.055	0.08 ± 0.04
N_5	0.033	0.04 ± 0.03
O_1	0.90	
O_2	0.60	0.69 ± 0.08
O_3	0.68	0.67 ± 0.07
O_4	0.010	
O_5	0.005	
P_1	0.176	
P_2	0.095	
P_3	0.110	
Q_1	0.016	
	45.9	

same result, namely $(0.30 \pm 0.02)\%$ $E2$.

Attempts to find the total 7.86-keV intensity and to compare it with the known population of the state by electromagnetic transitions (and thereby deduce possible electron capture to the state) are difficult not only because the O_1 and P shell lines are obscured by L Auger lines but also because the M lines are too low in energy for reliable intensity measurements. We have, however, made an estimate using the calculated internal-conversion coefficients of Dragoun *et al.*[27] for N, O, P, and Q shells and the Hager and Seltzer[28] calculations for the M shells. We extrapolate on a log-log plot of conversion coefficient vs energy N, O, P, and Q and use the computer interpolation suggested by Hager and Seltzer[28] for the M shells. Table V gives the calculated intensities for 0.997 $M1 + 0.003$ $E2$ normalized on the N_{1-3} average experimental intensities. The total intensity thus derived is *smaller* by ~20% than the sum of the incoming transitions to the 7.86-keV state. While this discrepancy is at the limit of our experimental uncertainty concerning the intensity of the N and O lines, it is also true that no other test of the M/N conversion ratios for these calculations at such a low energy is known to us.

In Table III the intensities are given in percent per decay on the assumption that the E.C. feed to the ground-state band is 10%. The γ intensities and the electron intensities are related through

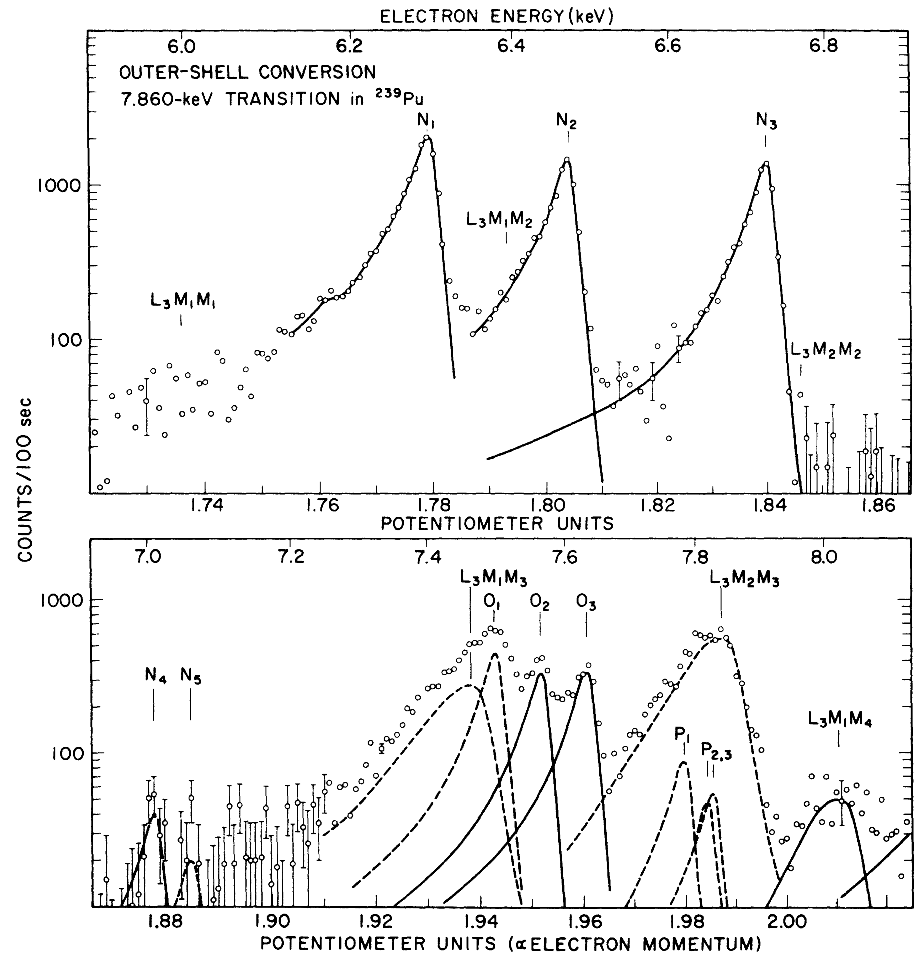

FIG. 5. Outer-shell conversion lines of the 7.860-keV transition in ^{239}Pu. The abscissa is a logarithmic momentum scale. We used the N_3 line shape for the analysis of the O and P lines. The dashed curves for O_1, N_5, and P lines have the intensities predicted by theoretical conversion coefficients and by the $M1$-$E2$ mixing derived from the N-subshell intensities (see Table V). All the L Auger lines expected in this region are indicated. The much broader line shape for the L Auger lines is obtained from the well-resolved $L_3M_3M_3$ line at 8.8 keV (not shown). A background of 210 counts/100 sec has been subtracted from all data points. Possible structure on the tail of the N_1 line at 1.762 potentiometer units may be due to "shakeoff" of $O_{4,5}$ electrons accompanying N_1 conversion.

the e_K/γ ratio of the 277.6-keV transition as measured with the solid-state detectors (Sec. III C). In addition to the electron lines reported here, about 50 lines (including many multiplets) of the L Auger spectrum were observed. These will be reported elsewhere.

C. Electron Spectroscopy with Cooled Si(Li) Detector

The conversion-electron spectrum of the ^{239}Am sample was also measured with a 80-mm^2×3-mm Si(Li) detector. The detector was cooled to liquid-nitrogen temperature and coupled to a low-noise preamplifier. The electron spectrometer had a resolution (FWHM) of 1.2 keV at 100 keV and 1.7 keV at 600-keV electron energy. The detector had a slightly different response to electrons compared with its response to γ rays. At 100 keV the electron energy was lowered by 0.7 keV with respect to that of a γ ray. This is due to the loss of electron energy in the detector window.

The electron spectrum of a mass-separated ^{239}Am sample measured with the cooled Si(Li) detector is shown in Fig. 6. The absolute K conversion coefficient of the 277.6-keV transition was measured with respect to the K conversion coefficient of the 279.2-keV transition in ^{203}Tl, which is known very accurately[29] (0.163 ± 0.002). The γ-ray and conversion-electron spectra of ^{239}Am and ^{203}Hg samples were measured at identical geometries. Since the γ and e^- spectra were measured at different times, decay corrections were applied to the spectra. These measurements gave a value of 1.11 ± 0.05 for the K conversion coefficient of the 277.6-keV transition.

The electron intensities and conversion coefficients obtained from the Si(Li) spectra are given in Table VI. Intensities are expressed in percent per ^{239}Am E.C. decay. The conversion coefficients for the transitions above 100 keV are more accurately determined; the comparatively larger errors on the lower-energy transitions are due to larger error in the γ-ray intensities and to larger uncertainties in the electron-detection efficiencies. Energy-independent efficiency was assumed in the energy range presented. Comparison with magnetic spectrometer results showed agreement within the errors assigned.

An attempt was made to determine the E.C. feed to the 57.3-keV level of ^{239}Pu. Electron spectra of mass-separated ^{239}Am and ^{239}Np samples were measured with a cooled Si(Li) detector at identical geometries. Since the main γ-ray population into the ground-state band and the intraband transition ratios are quite similar in the decay schemes of ^{239}Am and ^{239}Np, the ratio of the intensities of 277.6- and 57.3-keV transitions should be the same if the $\frac{5}{2}$ and higher members of the ground-state band receive the same amount of direct population in ^{239}Am and ^{239}Np decays. The ratios of the 57.3-keV (L_1+L_2) to the 277.6-keV K electron intensities were determined to be 0.695 and 0.690 for ^{239}Am and ^{239}Np, respectively. These ratios give a value of (3.0 ± 1.5)% for the E.C. feed to the 57.3- and 75.7-keV levels, based on a 4% ^{239}Np β^-

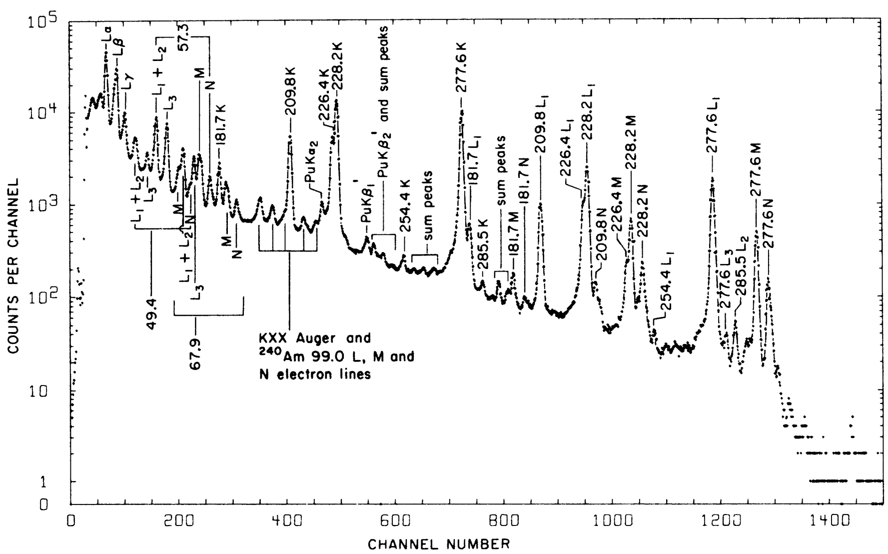

FIG. 6. Conversion-electron spectrum of a mass-separated ^{239}Am sample measured with a cooled Si(Li) detector. Sum peaks with L x rays appear emphasized in the spectrum because (intense) e^- lines are in cascade with Pu L x rays; the summing occurs in spite of the low-geometry-efficiency product of 1% for the peak areas.

feed[30] to these same levels of ^{239}Pu. In deducing this E.C. feed we have assumed that 5% of the 57.3-keV (L_1+L_2) is contributed by the upper 57.3-keV transition.

D. Half-Life and $\alpha/(\alpha+$E.C.) Branching Ratio

The half-life of the ^{239}Am was measured by following the decay of the 277.6-keV γ ray. The half-life was found to be 11.9 ± 0.1 h, in good agreement with the value (12.1 ± 0.4 h) reported by GCG.[2] The α-particle energy of the most prominent group was measured to be 5.772 ± 0.005 MeV, in agreement with the recently measured value[31] (5.776 ± 0.002 MeV). The $\alpha/(\alpha+$E.C.) ratio was determined to be $(1.0\pm0.1)\times10^{-2}$%, using the 277.6-keV intensity as 15 photons per 100 E.C. decays.

TABLE VI. ^{239}Am electron data obtained with the cooled Si(Li) detector.

Transition (keV)		Intensity (% per decay)	Conversion coefficient [a]	Multipolarity [b]
49.4	(L_1+L_2)	5.7	54 ± 5	
	L_3	2.4	23 ± 2	$M1+E2$
	M	2.4	23 ± 2	
	N	0.66	6.2 ± 0.7	
57.3	(L_1+L_2)	11.7	71 ± 7	$E2$
	L_3	11.0	66 ± 6	
	M	6.2	37 ± 3	
	N	2.0	12 ± 1.2	
67.8	(L_1+L_2)	4.8	38 ± 4	$E2$
	L_3	3.8	30 ± 3	
	M	2.1	16.7 ± 1.7	
	N	0.83	6.5 ± 0.7	
181.7	K	3.1	2.9 ± 0.2	$M1$
	(L_1+L_2)	0.81	0.75 ± 0.06	
	M	0.20	0.18 ± 0.02	
	N	0.063	0.058 ± 0.006	
209.8	K	8.0	2.3 ± 0.15	$M1$
	L_1+L_2	1.7	0.47 ± 0.03	
	N	0.15	0.043 ± 0.004	
226.4	K	5.9	1.8 ± 0.13	$M1$
	(L_1+L_2)	1.4	0.41 ± 0.03	
	M	0.38	0.11 ± 0.01	
	N	0.14	0.040 ± 0.004	
228.2	K	21.6	1.9 ± 0.12	$M1$
	L_1+L_2	4.5	0.40 ± 0.03	
	M	1.0	0.09 ± 0.01	
	N	0.35	0.031 ± 0.003	
254.4	K	0.15	1.8 ± 0.2	$M1$
	L_1+L_2	0.029	0.34 ± 0.04	
277.6	K	16.7	1.11 ± 0.05	$M1$
	L_1+L_2	3.41	0.23 ± 0.015	
	L_3	0.036	0.0024 ± 0.0002	
	M	0.86	0.057 ± 0.004	
	N	0.29	0.019 ± 0.002	
285.5	K	0.075	0.094 ± 0.009	$E2$
	L_1+L_2	0.065	0.08 ± 0.008	

[a] Si(Li) and Ge(Li) detector system calibrated with the 279-keV transition in Tl^{203}; K conversion coefficient = 0.163 ± 0.002.

[b] The $E2$ admixture in the lower-energy transitions is more accurately determined by the magnetic spectrometer and is given in Table III.

IV. DISCUSSION

A. Decay Scheme

The decay scheme of ^{239}Am is shown in Fig. 7. The assignments of the ground state, 285.46-, and 391.6-keV levels to the $\frac{1}{2}$+(631), $\frac{5}{2}$+(622), and $\frac{7}{2}$−(743) Nilsson states[7] have been made previously from the study of the β^- decay[3,4] of ^{239}Np and α decay[5,6] of ^{243}Cm. The ground-state spin has also been determined experimentally[32] to be $\frac{1}{2}$. The other assignments are based mainly on the measured multipolarities of the transitions orginating from these levels. The results of the present investigation fully support the above assignments. From the observed energies of the members of the ground-state $\frac{1}{2}$+(631) band we calculate its rotational constant, $\hbar^2/2\mathscr{I}$, and decoupling parameter a, as 6.251 keV and −0.581, respectively. The rotational constant of the $\frac{5}{2}$+(622) band is calculated to be 6.380 keV.

The level at 511.8 keV has been found to decay to the $\frac{5}{2}$, $\frac{7}{2}$, and $\frac{9}{2}$ members of the $\frac{5}{2}$+(622) band. The $M1$ multipolarities of these three transitions (226.4, 181.7, and 124.4 keV) uniquely determine the spin and parity of the 511.8-keV state as $\frac{7}{2}$+. This level is therefore given an assignment of $\frac{7}{2}$+(624), which is the only $\frac{7}{2}$+ single-particle state expected in this energy region.

The weak 430.0- and 497.8-keV γ rays, as indicated in Fig. 7, orginate from a level at 505.7 ± 0.2 keV. This deexcitation pattern to the ground-state band is quite similar to that of a 505.3 ± 0.2-keV level populated in the ^{239}Np decay,[8] namely, the transition to the 57.27-keV level is relatively weak compared with the transitions to the 75.70- and 7.86-keV levels. We equate these levels and use the Davies and Hollander[8] deduction that it is the spin-$\frac{5}{2}$ member of the $\frac{1}{2}$−(501) band. There is, however, an obvious difference in the β decay transitions to the band from the two parent states. The ^{239}Np β^- decay favors the spin-$\frac{3}{2}$ member of the band, whereas the ^{239}Am electron capture favors the spin-$\frac{5}{2}$ member.

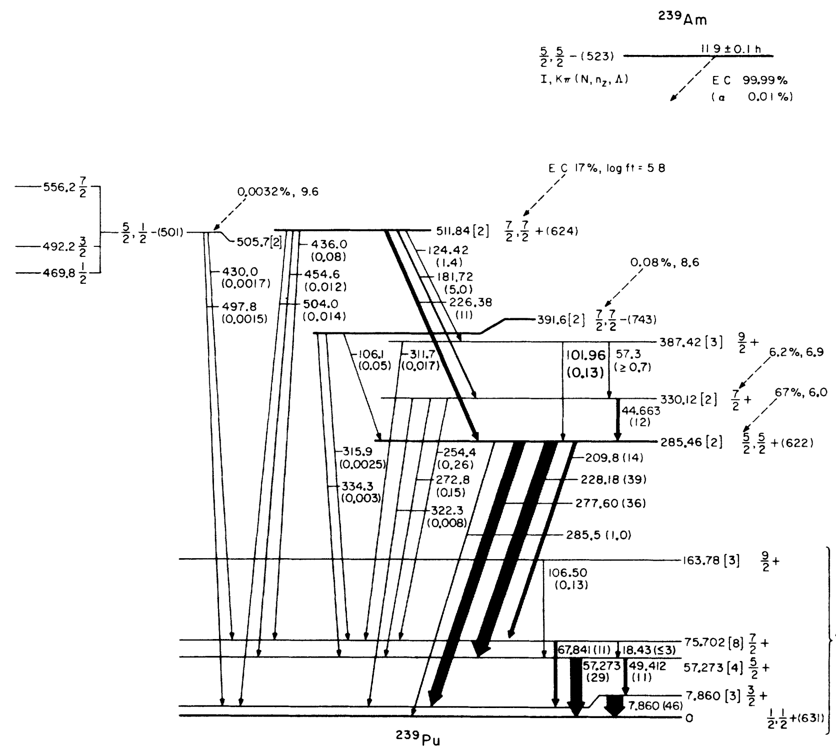

FIG. 7. Decay scheme of ^{239}Am to levels in ^{239}Pu. Energies are in keV. The only energy errors given here are for the level energies; the error in the last quoted figure is given in square brackets [ ]. Transition-energy errors are listed in Tables I and III. Transition intensities are given in parentheses near the transition energy, units are (percent per ^{239}Am decay). The spin, parity, and quantum-number assignments for the 511.8-keV level and the multipolarities of the new low-intensity deexcitations of the 387.4-keV state are from present work. Transition intensities have errors between 10–20% plus a correlated contribution from the uncertainty in the assigned 10% electron capture to the ground-state band. At the left are other members of the $\frac{1}{2}$ − (501) band not observed in ^{239}Am decay but seen in the decay of ^{239}Np (Ref. 8).

B. Electron-Capture Transition Probabilities

The present experiments do not yield any direct information about the E.C. feed to the $\frac{1}{2}$ and $\frac{3}{2}$ members of the ground-state band. However, the ratios of the 57.3-keV (L_1+L_2) and 277.6-keV K electron intensities in the ^{239}Np and ^{239}Am electron spectra indicate that the 57.3-keV level of ^{239}Pu receives almost equal amounts of direct population in the decay of both nuclides. In the ^{239}Np decay the β^- decay intensity to the ground-state band has been measured[30] to be 10%. We, therefore, have assigned 10% E.C. feed to the ground-state band. The E.C. feed to other levels was calculated by summing the intensities of γ rays and conversion electrons originating from that level. The intensities thus determined are given in Fig. 7. The log*ft* values were calculated by the method of Major and Biedenharn,[33] using 810 keV for the ^{239}Am decay energy.[34]

The very high log*ft* value of 8.6 for the E.C. transition to the $\frac{7}{2}-(743)$ state at 391.6 keV clearly shows that the ground state of ^{239}Am is different from that of ^{239}Np (the log*ft* value of β^- decay to this state of 6.5). The ground states of all of the known[35] odd-mass Am isotopes are found to be either the $\frac{5}{2}-(523)$ or the $\frac{5}{2}+(642)$ Nilsson state. Since the ground state of ^{239}Np is known[35] to be $\frac{5}{2}+(642)$, the ground state of ^{239}Am should be $\frac{5}{2}-(523)$. This assignment is in agreement with the assignment[36] made on the basis of α decay of ^{243}Bk.

The large retardation in the E.C. transition probability to the $\frac{7}{2}-(743)$ state is in agreement with the theoretical expectation. Recent calculations by Chasman[37] show that the $\frac{7}{2}-(743)$ state is 90% $J=\frac{15}{2}$ and the $\frac{5}{2}-(523)$ state is 90% $J=\frac{9}{2}$. The mismatch between the angular momenta of the two states causes the high retardation in E.C.-decay transitions.

The total number of K-shell holes per decay can be calculated from the theoretical prediction[38] of $K/$ total capture and the experimental number of K conversions per decay. For ^{239}Am we find 52% K holes per decay from K conversion and 70% K holes per decay from K capture. Using a K fluorescence yield of 97%, we predict 118 K x rays per 100 E.C. decays, which is in agreement with the experimental result of 116 ± 4 K x rays per 100 ^{239}Am E.C. decays. This agreement is additional support for the assignment of ~10% E.C. population of the ground-state band.

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Properties of Levels in ^{198}Pb and ^{200}Pb†

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The decay of ^{198}Bi and ^{200}Bi to levels in the corresponding Pb daughters have been investigated using sources which were both mass-separated and chemically processed. γ-ray and conversion-electron singles spectra were obtained, as well as γ-γ prompt- and delayed-coincidence spectra. These data yield levels in ^{198}Pb at the following energies (spin-parities): 1062.9 (2^+), 1624.9 (4^+), 1822.3 [$(5)^-$], and 2139.8 [$(7)^-$] keV. Those in ^{200}Pb are at 1026.3 (2^+), 1488.3 (4^+), 1907.9 [$(5)^-$], and 2152.9 [$(7)^-$) keV. The half-life of the 1822-keV level in ^{198}Pb was determined to be 63.3± 2.5 nsec, and those of the 1908- and 2153-keV levels in ^{200}Pb are 1.50± 0.08 and 47.6± 2.5 nsec, respectively.

I. INTRODUCTION

Recent investigations[1-3] of the neutron-deficient isotopes $^{192-200}$Hg have given new information about the lowest collective excitations of these even nuclei and have also revealed new negative-parity levels. The properties of the collective positive-parity states show a remarkable independence of the neutron number and appear to have characteristics midway between those expected for a vibrating spherical nucleus and a rigid rotating spheroidal nucleus. The properties of the negative-parity states, however, appear to be more dependent on neutron number. This is rather surprising in view of their theoretical description[4,5] as two-proton-hole states coupled to phonon excitations. In order to study further the nuclear level structure in this region, we have extended our investigations to the neutron-deficient Pb isotopes where proton shell closure occurs.

During the course of our experiments, which we have reported in part earlier,[6] we learned of similar investigations by Hanser[7,8] and by Ishihara.[9] The results of these authors appear part-