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ABSTRACT

The conversion electron spectra arising from electron-capture decay of Am^{239} and Am^{240} have been studied with a 180° photographic spectrograph at ~0.1 percent resolution. The Am^{239} spectrum and the multipolarity data obtained therefrom are similar to those observed from the beta decay of Np^{239}, but the electron intensity data indicate somewhat different relative populations of the Pu^{239} excited states in the two cases. An attempt to assign the spin and parity of Am^{239} from ft values by the use of ΔI and ΔK selection rules was not successful.

The energies of the first two excited states of Pu^{240} are accurately measured, and from these values the constants in the Bohr-Mottelson two-term rotational formula are evaluated.

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INTRODUCTION

The level structure of the Pu^{239} nucleus has been defined as the result of several investigations of the Np^{239} beta decay,[1-3] the Cm^{243} alpha decay,[4, 5] and Pu^{239} Coulomb excitation.[6]

Two rotational bands in Pu^{239} are seen from Np^{239} beta decay,[2] an "anomalous" $K = \Omega = 1/2$ band based at the ground state and a "normal" $K = \Omega = 5/2$ band based at 286 kev; the ground-state band has also been observed[6] from Coulomb excitation of Pu^{239}. In addition to the rotational levels of these bands, an $I = 5/2$ or $7/2-$ state has been seen at 392 kev and an $I = 5/2$ or $7/2+$ state is found at 512 kev.

In the interpretation of the beta decay groups from Np^{239} serious difficulty is encountered in attempting to reconcile the experimental log ft values with the measured[7] spin of 1/2 for Np^{239}. These difficulties may be summarized as:

1) Beta transitions to levels of the ground-state rotational band are very slow (log ft ≥ 9), whereas the ground-state to ground-state beta transition is expected to be in the allowed ($\Delta I = 0$, no) or first forbidden ($\Delta I = 0$, yes) classification.

2) Beta transitions take place to levels of 5/2+ (286 kev) and 5/2- (392 kev) with log ft 7.0 and 6.5 respectively. This is clearly an inconsistency with fundamental beta-decay selection rules, since one or the other of these two transitions would have to be in the second forbidden ($\Delta I = 2$, no) classification and would be unobservable.

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If the interpretation of the Pu^{239} level scheme as proposed by Newton et al.[5] and HSM[2] is correct, the spin of Np^{239} cannot be 1/2. The latter group of investigators suggest that the spin of Np^{239} may be 3/2 or 5/2, with 5/2 preferred because such an assignment would make all beta transitions to the ground-state rotational band K-forbidden and hence slow.

The electron-capture decay of the 12-hr isotope Am^{239} also populates the excited states of Pu^{239}, but the decay properties of this isotope had not previously been studied using techniques of high-resolution beta spectroscopy. In the interest of examining the energy levels of Pu^{239} populated from the electron-capture side and hence forming a more nearly complete picture of all radioactive decays leading to levels of Pu^{239}, and also in an attempt to shed light upon the nature of the beta decay processes occurring here, we have prepared Am^{239} and studied its conversion electron spectrum at ~ 0.1 percent resolution using the Berkeley permanent-magnet beta spectrographs.[8]

The sample of Am^{239} used in these experiments was prepared by a (d, 2n) reaction upon Pu^{239}, using 20-Mev deuterons from the Crocker 60-inch cyclotron. Since the longer-lived (~ 50 hr) isotope Am^{240} is also produced under these conditions by the (d, n) reaction, it was also possible to study its conversion electron spectrum as the Am^{239} decayed and hence to examine at high precision the energy levels of the even-even isotope Pu^{240}. The first excited state of Pu^{240} had previously been measured[8] as 42.88 ± 0.05 kev from alpha decay of Cm^{244}.

II. EXPERIMENTAL PROCEDURE

One of the flat 180° permanent-magnet beta spectrographs, of effective field strength 99 gauss, was used to record photographically the conversion lines upon glass-backed Eastman No-Screen X-ray plates. A description of these instruments and of their calibration has been given by Smith and Hollander.[8] In addition to the previous calibration, a small amount of Am^{241} was introduced into the sample to serve conveniently as an internal standard, since the lines of the Am^{241} gamma rays (whose energies have been determined to better than 0.1 percent by Day[9]) become predominant as the Am^{239} and Am^{240} decay. As a result of this internal calibration, we feel that the values of the low-energy transitions obtained in the present study should supersede those reported from the previous study[2] of Np^{239} (although in no case do the results deviate by as much as 0.2 percent).

The americium fraction was purified from the target material and fission products by the following procedure: The PuO_2 target was dissolved in nitric plus hydrochloric acids, and lanthanum added as a carrier for americium. After separating the Am(III) and rare-earths from Pu(IV) by means of a Dowex A-1 anion exchange column at ~13 M HCl, LaF_3 and $La(OH)_3$ precipitations were made as purification steps. Americium was then separated from the lanthanum carrier by means of an alcoholic-HCl Dowex-50 cation-exchange column according to the procedure of Thompson, et al.[10]

The hydrochloric acid solution was evaporated to dryness and the americium activity dissolved in 0.5 ml of NH_4HSO_4 plating solution,[8] from which the activity was electroplated upon a 10-mil platinum wire and then placed into the spectrograph camera.

III. EXPERIMENTAL RESULTS, Am^{239}

The conversion electron data are summarized in Table I; measured electron energies are given, followed by the shell or subshell assignment, the transition energy, and the visual intensity estimate. The transition energy selected is a weighted average based on line intensity and proximity of lines to calibration points. Because the sample of Am^{239} used in this experiment was much weaker than the Np^{239} samples used by HSM,[2] fewer lines were seen and these were less intense. However, the present data have indicated certain definite differences in population of Pu^{239} levels between Am^{239} decay and Np^{239} decay and therefore they provide new information about the beta-decay processes involved.

The total transition intensities are summarized in Table II. The electron intensities are those obtained in the present experiment, but most of the multipole orders and mixing ratios have been taken from HSM.[2] For comparison, the transition intensities obtained by HSM from Np^{239} decay are also included in Table II.

Several facts may immediately be observed from Table II:

a) The 181.8-and 226.5-kev transitions, only very weakly excited in Np^{239} decay, are more abundantly in evidence from Am^{239} decay. These transitions depopulate the 512.3-kev state in Pu^{239}.

Table I. Conversion electron data for Am^{239}

Electron Energy (kev)	Shell	Transition Energy (kev)	Visual Intensity Estimate	Intensity (Densitometer)	Remarks
21.63	L_I	44.73	vw		
22.41	L_{II}	44.66	vvw		$E\gamma$ = 44.70 kev
23.10	L_I	49.46	w		
27.20	L_{II}	49.47	w-m	40	
31.41	L_{III}	49.47	w-m	40	
43.48	M_I	49.41	vvw		
43.94	M_{II}	49.50	w		
44.98	M_{III}	49.54	vw-w		
48.10	N	~49.4	vvw broad		$E\gamma$ = 49.47 kev
34.17	L_I	57.27	vvw		
35.05	L_{II}	57.30	m-s	200	
39.29	L_{III}	57.35	m-s	200	
51.78	M_{II}	57.34	m	90	
52.75	M_{III}	57.31	m	80	
55.91	N_{II}	57.29	w		
56.20	N_{III}	57.33	vw		
57.06	O	57.3	vw		$E\gamma$ = 57.31 kev
45.69	L_{II}	67.94	ms	107	
49.43	L_{III}	67.91	m	98	
62.35	M_{II}	67.91	w-m		
63.34	M_{III}	67.90	w-m		
66.79	N	~68.0	w broad		
67.81	O	~68.0	vw broad		$E\gamma$ = 67.91 kev
59.92	K	181.7	m	77	
158.8	L_I	181.9	w		$E\gamma$ = 181.8 kev

Electron Energy (kev)	Shell	Transition Energy (kev)	Visual Intensity Estimate	Intensity (Densitometer)	Remarks
87.92	K	209.7	s	240	
187.0	L_I	210.1	wm		
187.7	L_{II}	210.0	vw		
204.1	M_I	210.0	vw		Eγ = 209.9 kev
104.6	K	226.4	ms	190	
203.6	L_I	226.7	w		
220.7	M_I	226.6	vw		Eγ = 226.5 kev
106.4	K	228.2	vs	640	
205.3	L_I	228.4	m		
206.1	L_{II}	228.4	w		
222.4	M_I	228.3	w		
226.8	N_I	228.4	vvw		Eγ = 228.3 kev
155.9	K	277.7	s	670	
254.5	L_I	277.6	m		
255.3	L_{II}	277.6	vw		
271.6	M_I	277.5	vw		Eγ = 277.6 kev

s = strong, m = moderate, w = weak, v = very

Table II. Transition data.

Transition Energy (kev)	Total Electron Intensity$^{(a,b)}$	Multipole Order$^{(c)}$	Conversion Coefficient$^{(e)}$	Transition Intensity from Am^{239} $N_e + N_\gamma$ $^{(b)}$	Transition Intensity$^{(2)}$ from Np^{239} $N_e + N_\gamma$ $^{(b)}$
44.70	$\leq$ 300	M1(80%)+E2(20%)$^{(d)}$	large (β_L = 75)	weak	~300
49.47	~ 350	M1(70%)+E2(30%)$^{(d)}$	large (β_L = 55)	~350	475
57.31	1275	E2(M1<5%)$^{(d)}$	large (α_L = 190)	1275	1275
61.4	--	-------	------	--	~350
67.91	660	E2 (predominantly)$^{(d)}$	large (α_L = 85)	660	800
106.1	--	-------	------	--	2300
181.8	230	M1(E2<50%)	(β_Σ = 7.0)	260	weak
209.9	710$^{(f)}$	M1(E2<30%)$^{(d)}$	(β_Σ = 5.0)	850	840
226.5	570	M1(E2<40%)	(β_Σ = 3.8)	720	weak
228.3	1980	M1(E2<20%)$^{(d)}$	(β_Σ = 3.8)	2500	2420
277.6	1420$^{(f)}$	M1(E2<10%)$^{(d)}$	(β_Σ = 2.2)	2050	2000

a. We have taken $L/(M+N) \approx 3$.

b. Normalized to $I_{57.31} = 1275$

c. Mixing ratios have been obtained from L-subshell internal conversion data.

d. Obtained from HSM.[2]

e. All conversion coefficients in parentheses are the theoretical values of Rose.[11]

f. Assume $K_{210}/K_{228}/K_{278} = 1.0/2.8/2.0$.

b) The 61.4- and 106.1-kev electric dipole transitions observed in Np^{239} decay are absent in Am^{239} decay. In the electron plates from Np^{239} decay, the L-subshell conversion lines of the 106-kev transition had appeared with comparable intensity to the L-lines of the 49.4-kev transition or the M-lines of the 67.8-kev transition; hence from their absence in the Am^{239} plates one can set an upper limit on the population of the 392-kev state by electron capture of Am^{239}. Such a limit is found to be ~2 percent.

c) There are no new levels populated from Am^{239} decay which have not been seen from Np^{239} decay.

The level scheme of Pu^{239}, from the combined data of HSM and the present paper, is shown in Fig. 1. Also included are the Np^{239} beta and Am^{239} electron-capture branches and their log ft values. (The electron-capture disintegration energy has been estimated from the thermodynamic data of Glass, Thompson, and Seaborg.[12]).

IV. INTERPRETATION OF RESULTS

The amount of electron-capture branching of Am^{239} to the ground-state rotational band is not known, so one can actually calculate only lower limits to the ft values for decay to the other states. However, the intensity figures for the 57.3- and 67.9-kev gamma rays indicate that there is little if any direct population to the $I = 5/2$ and $I = 7/2$ states of this band, so perhaps the quoted ft values are not too inaccurate.

The log ft for the electron capture transition to the 286-kev state ($I = 5/2+$) is ≥ 5.9, which would indicate either an allowed or first forbidden transition with $\Delta I = 0, 1$. The spin of Am^{239} would then be 3/2, 5/2, 7/2±. The fact that the log ft value for the transition to the 392-kev state (5/2, 7/2-) is greater than eight would indicate $\Delta I = 2$, yes or no, and that the spin of Am^{239} could be 1/2 or 3/2 (or 9/2, 11/2). From the two pieces of information it would appear that the spin of Am^{239} should be 3/2. However, such a spin would be inconsistent with the observation that there is little or no electron capture to the 57.3-kev state (5/2+). Thus, the ΔI selection rules alone do not seem to allow a consistent explanation to be given of the electron-capture branching of Am^{239}.

The spin of Am^{241} has been measured[13] as 5/2 and it would not be unreasonable that Am^{239} should also have spin 5/2 (especially in view of the similarities in alpha-decay properties of Am^{239}, Am^{241}, and Am^{243}).[14] Such an assignment would allow an explanation of the slowness of decay to the ground-state ($K = 1/2$) band by means of the K selection rule, which prohibits transitions where ΔK exceeds the multipolarity L. (In such a case, a $\Delta I \geq 2$ beta transition could proceed normally.) A spin of 5/2 for Am^{239}, however, cannot account for the slowness of transitions to the 392-kev state (5/2, 7/2-) either with ΔI or ΔK selection rules.

In the following paper, a possible explanation for the beta-decay branchings of Np^{239} and Am^{239} is given which makes use of the recent theoretical results of Nilsson[15] and Alaga.[16]

V. EXPERIMENTAL RESULTS, Am^{240}

Am^{240} decays by electron capture to Pu^{240} with a half-life of about two days.[17,18] Glass[19] has reported gamma rays of 0.92, 1.02, and 1.40 Mev in the scintillation spectrum of this isotope, but the conversion electron spectrum had not been studied previously. We report here measurements of the energies of the $4+ \rightarrow 2+ \rightarrow 0+$ (gd state) gamma-ray cascade in Pu^{240} following electron capture of Am^{240}.

Table III summarizes the Am^{240} electron energy data. The energy of the first excited state of Pu^{240} as determined from M_{II} and M_{III} conversion lines is 42.87 kev; the accuracy of this value should be better than 0.1 percent since these two lines lie extremely close to the L_I and L_{II} lines of the 59.57 ± 0.02-kev gamma ray of Am^{241}. This Pu^{240} energy had previously been measured from the Cm^{244} electron spectrum[8] as 42.88 ± 0.05 kev.

The energy of the E2 transition from the 4+ to the 2+ state is 98.90 ± 0.2 kev; thus the energy of the second excited state of Pu^{240} is 141.77 ± 0.2 kev. From these energies one can calculate the constants in the Bohr-Mottelson equation for rotational states of a deformed nucleus:

$$E_i = A\, I(I+1) - B\, I^2(I^2+1),$$

where I = spin of state i,

$A = \hbar^2/2\mathfrak{J}$,

$B = 2(1/\hbar\omega)^2 \; (\hbar^2/\mathfrak{J})^3$,

$\mathfrak{J}$ = nuclear moment of inertia,

$\hbar\omega$ = vibrational quantum energy.

Table III. Conversion electron data for Am^{240}.

Electron Energy (kev)	Shell	Transition Energy (kev)	Visual Intensity Estimate	
20.63(a)	L_{II}	42.88	m	
24.82(a)	L_{III}	42.88	m	
(37.16)(a)	(L_I Am^{241})	(59.57)	(m)	
37.30	M_{II}	42.86	w	
(37.98)(a)	(L_{II} Am^{241})	(59.57)	(m-s)	
38.32	M_{III}	42.88	w	
41.54	N	~42.8	w	
42.66	O	~42.9	vvw	$E\gamma = 42.87 \pm 0.03$ kev
76.63	L_{II}	98.88	m-s	
80.83	L_{III}	98.89	m	
93.35	M_{II}	98.91	w-m	
94.37	M_{III}	98.93	w-m	
97.74	N	~99.0	w broad	
98.64	O	~98.8	vvw	$E\gamma = 98.90 \pm 0.2$ kev

a. Assumed energy.

Such a calculation yields A = 7.16 ± 0.01 kev and B = 0.005 ± 0.002 kev. These results may be compared with the values obtained from the energies of Pu^{238} excited states.[8] A = 7.37 ± 0.01 kev and B = 0.005 ± 0.003 kev.

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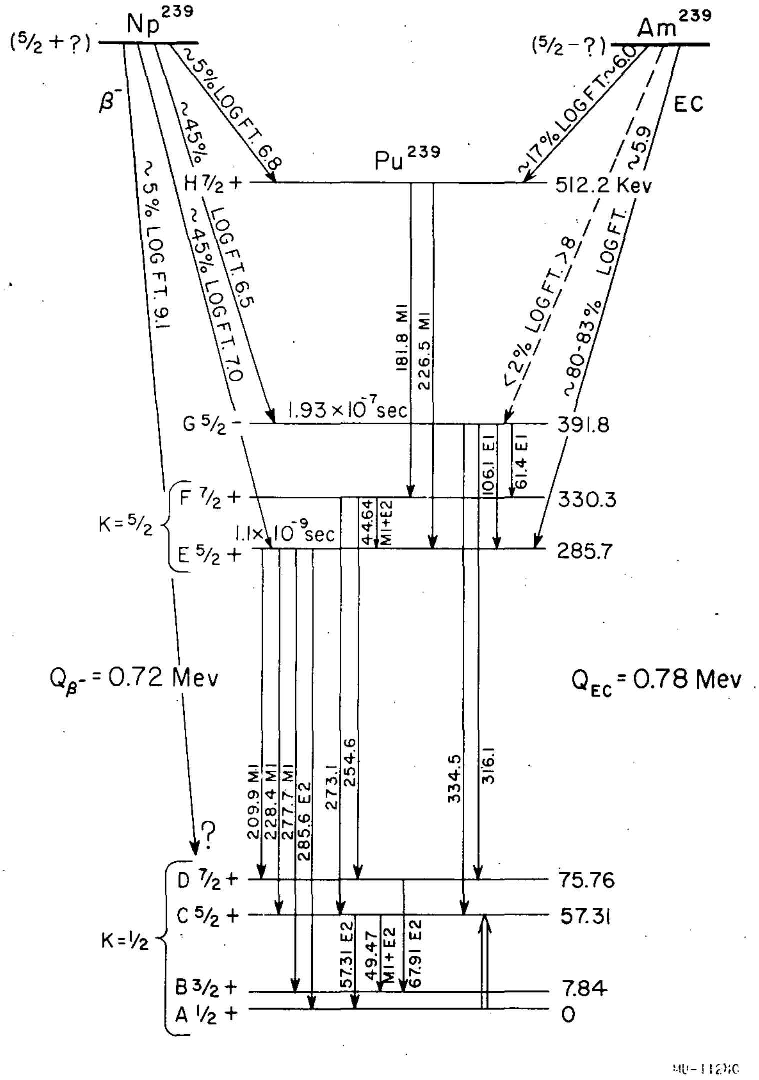

Fig. 1. Level scheme of Pu^{239}.