Electron-Capture Decay of Am^{239} and Am^{240}†

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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% 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 does not give a consistent explanation.

The energies of the first two excited states of Pu^{240} are accurately measured, and from these values (42.87±0.04 and 141.77±0.2 kev) the constants in the Bohr-Mottelson two-term rotational formula are evaluated: $A=7.16\pm0.01$ kev, $B=0.005\pm0.002$ kev.

I. 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 difficulties are 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 follows:

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

(2) Beta transitions are observed to take place to levels of $5/2+$ (286 kev) and $5/2-$ (393 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 have a very long lifetime. If the interpretation of the Pu^{239} level scheme as proposed by Newton *et al.*[5,6] 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 by means of 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% 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

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[1] Freedman, Wagner, Engelkemeir, Huizenga, and Magnusson (private communication, November, 1952).
[2] Hollander, Smith, and Mihelich, Phys. Rev. **102**, 740 (1956). This paper will hereafter be referred to as HSM.
[3] D. Engelkemeir and L. B. Magnusson, Phys. Rev. **99**, 135 (1955).
[4] I. Perlman and F. Asaro, *Annual Review of Nuclear Science* (Annual Reviews, Inc., Stanford, 1954), Vol. 4, p. 157.
[5] Newton, Rose, and Milsted, Phil. Mag. (to be published).
[6] J. O. Newton, Nuclear Phys. (to be published).
[7] J. G. Conway and R. D. McLaughlin, Phys. Rev. **96**, 541 (1954).
[8] W. G. Smith and J. M. Hollander, Phys. Rev. **101**, 746 (1956).

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% 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%).

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 ~13M 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.

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

Electron energy (kev)	Shell	Transition energy (kev)	Visual intensity estimate[a]	Intensity (densitometer)	E_γ (kev)
21.63	L_I	44.73	vw		
22.41	L_{II}	44.66	vvw		44.70
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		49.47
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		57.31
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		67.91
59.92	K	181.7	m	77	
158.8	L_I	181.9	w		181.8
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		209.9
104.6	K	226.4	ms	190	
203.6	L_I	226.7	w		
220.7	M_I	226.6	vw		226.5
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		228.3
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		277.6

[a] s ≡ strong, m ≡ moderate, w ≡ weak, v ≡ very.

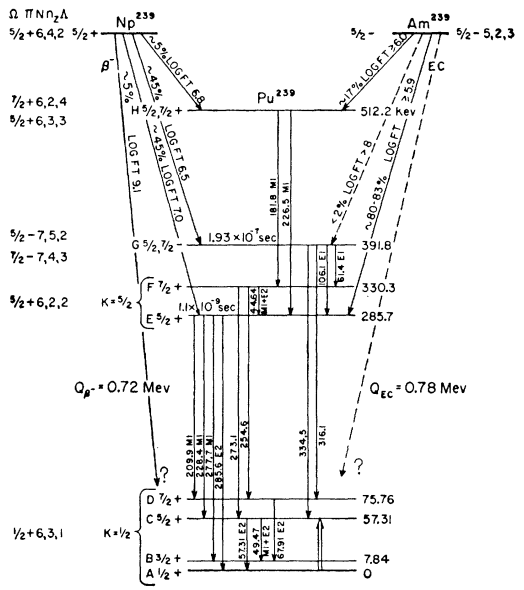

FIG. 1. Level scheme of Pu^{239}.

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 new information is provided 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]

[9] P. P. Day, Phys. Rev. **97**, 689 (1955).

[10] Thompson, Harvey, Choppin, and Seaborg, J. Am. Chem. Soc. **76**, 6229 (1954).

TABLE II. Transition data.

Transition energy (kev)	Total electron intensity[a,b]	Multipole order[c]	Conversion coefficient[d]	Transition intensity from Am^{239} N_e+N_γ[b]	Transition intensity from Np^{239} N_e+N_γ[b,e]
44.70	≤ 320	$M1(90\%)+E2(10\%)$	large ($\beta_L=45$)	≤ 320	~ 300
49.47	~ 350	$M1(80\%)+E2(20\%)$	large ($\beta_{LI}=31$)	~ 350	475
57.31	1275	$E2(M1<5\%)$	large ($\alpha_L=190$)	1275	1275
61.4	...	...	...	...	~ 350
67.91	700	$E2$ (predominantly)[c]	large ($\alpha_L=85$)	700	800
106.1	...	...	...	...	2300
181.8	230	$M1(E2<50\%)$	($\beta_\Sigma=5.2$)[h]	275	weak
209.9	710[f]	$M1(E2<30\%)$[c]	($\beta_\Sigma=3.3$)	920	910[g]
226.5	593	$M1(E2<40\%)$	($\beta_\Sigma=2.7$)	810	weak
228.3	1990	$M1(E2<20\%)$[c]	($\beta_\Sigma=2.7$)	2700	2600[g]
277.6	1400[f]	$M1(E2<10\%)$[c]	($\beta_\Sigma=1.5$)	2300	2200[g]

[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 of HSM.
[d] Conversion coefficients in parentheses are estimates based on the theoretical values of M. E. Rose [*Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), Appendix IV] and of L. A. Sliv [privately circulated tables (unpublished)]. An estimated finite size correction factor of 0.6 has been applied to the theoretical magnetic dipole coefficients for the L-shell.
[e] Obtained from HSM.
[f] Assume K_{210}: K_{228}: $K_{278}=1.0:2.8:2.0$.
[g] Slightly different from values in HSM because of the use of the corrected conversion coefficients.
[h] β_Σ=total $M1$ conversion coefficient.

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 512.2-kev state in Pu^{239}.

(*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 $\sim 2\%$.

(*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 logft values. (The electron-capture disintegration energy has been estimated from the thermodynamic data of Glass, Thompson, and Seaborg.[11])

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.

[11] Glass, Thompson, and Seaborg, J. Inorg. and Nuclear Chem. **1**, 3 (1955).

The logft 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, $7/2\pm$. The fact that the logft value for the transition to the 392-kev state (5/2, 7/2−) is greater than eight would indicate that $\Delta I=2$, 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 self-consistent explanation to be given of the electron-capture branching of Am^{239}.

The spin of Am^{241} has been measured[12] 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}).[13] 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 the transition 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[14] and Alaga.[15]

[12] M. S. Fred and F. S. Tomkins, Phys. Rev. **89**, 318 (1953).
[13] F. S. Stephens, University of California Radiation Laboratory Report UCRL-2970, 1955 (unpublished).
[14] S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **29**, No. 16 (1955).
[15] G. Alaga, Phys. Rev. **100**, 432 (1955).

IV. EXPERIMENTAL RESULTS, Am^{240}

Am^{240} decays by electron capture to Pu^{240} with a half-life of about two days.[16,17] Glass[18] has reported gamma rays of 0.92, 1.02, and 1.40 Mev in the scintillation spectrum of this isotope, but the low-energy conversion electron spectrum had not been studied previously. We report here measurements of the energies of the $4+ \rightarrow 2+ \rightarrow 0+$ (ground 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% since these two lines lie very 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:

$$E_i = AI(I+1) - BI^2(I^2+1),$$

where I = spin of state i, $A = \hbar^2/2\Im$, $B = 2(1/\hbar\omega)^2(\hbar^2/\Im)^3$, $\Im$ = nuclear moment of inertia, and $\hbar\omega$ = vibrational quantum energy. Such a calculation yields $A = 7.16 \pm 0.01$ kev and $B = 0.005 \pm 0.002$ kev. These results may be compared with the values obtained from the energies of Pu^{238} excited states[8]: $A = 7.37 \pm 0.01$ kev and $B = 0.005 \pm 0.003$ kev, and those from U^{234} excited states[19]: $A = 7.29 \pm 0.01$ kev and $B = 0.006 \pm 0.002$ kev.

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

Electron energy (kev)	Shell	Transition energy (kev)	Visual intensity estimate[a]	E_γ (kev)
20.62	L_{II}	42.87	m	
24.82	L_{III}	42.88	m	
(37.16)[b]	(L_I Am^{241})	(59.57)	(m)	
37.30	M_{II}	42.86	w	
(37.98)[b]	(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	42.87±0.03
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	98.90±0.2

[a] See reference *a* of Table I.
[b] Electrical energies calculated from transition energy 59.57 kev.

V. ACKNOWLEDGMENTS

We would like to thank Dr. R. A. Glass for his help with the chemical purification, and to acknowledge the help of the crew of the 60-inch cyclotron in performing the irradiation. The comments of Professor J. O. Rasmussen are also appreciated.

[16] Seaborg, James, and Morgan, *The Transuranium Elements: Research Papers* (McGraw-Hill Book Company, Inc., New York, 1949), National Nuclear Energy Series, Plutonium Project Record, Vol. 14B, Div. IV, p. 1525.
[17] G. H. Higgins, University of California Radiation Laboratory Report UCRL-1796, 1952 (unpublished).
[18] R. A. Glass, University of California Radiation Laboratory Report UCRL-2560, 1954 (unpublished).
[19] W. G. Smith and J. M. Hollander (unpublished results, 1955).