Electron-Capture Decay of Pu^{237}†

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The conversion and K-Auger electron spectra of Pu^{237} (produced by alpha bombardment of U^{235}) were examined with a permanent-magnet spectrograph and a solenoidal beta-ray spectrometer. The photon spectrum was studied with scintillation spectrometers and the following intensity ratios were determined: K x-radiation: L x-radiation: 59.6-kev gamma radiation = 1.00: 0.75: 0.14. Data from the beta-ray and scintillation spectrometers were used to calculate a K-Auger electron coefficient of 0.06 ± 0.01 for $Z = 93$. Coincidence measurements established the existence of both K- and L-electron capture to the 59.6-kev level of Np^{237}. The ratio of L to K capture to that level was found to be 2.8, and from this ratio a total electron-capture decay energy of 0.21 Mev was calculated for Pu^{237}. This value is in good agreement with that of 0.22 Mev estimated from closed decay-energy cycles. The experimental data indicate that the principal branches of electron-capture decay are to the 59.6-kev (15%) and the 33.2-kev (22%) levels, and to the ground state (60%) of Np^{237}, with corresponding logft values of 6.8, 6.9, and 6.8, respectively.

I. INTRODUCTION

PLUTONIUM-237 was first found in the products of helium ion bombardment of U^{235} and deuteron bombardment of Np^{237} by James and co-workers,[1,2] who reported that it decayed by electron capture with a half-life of about 40 days. This was later confirmed by Hoff[3] who observed the K and L x-rays of neptunium and a 64-kev gamma ray. Recently, a very low intensity alpha branch was detected in Pu^{237}.[4,5] Hoff reported that most of the electron capture in Pu^{237} proceeds to the 59.6-kev or lower levels of Np^{237}. He determined the following intensity ratios: K-capture: L-capture: electron-capture to the 59.6-kev level = 100: 67: 93. Since then, the levels of Np^{237} have been well established by studies of the alpha decay[6–10] of Am^{241} and the beta decay[11–14] of U^{237}. Therefore, a more thorough investigation of the decay of Pu^{237} was undertaken in order to determine the relative abundance of electron capture to the various levels of Np^{237}, the ratio of L- to K-electron capture, and if possible, the total electron-capture disintegration energy of Pu^{237}. The levels of Np^{237} at 33.2, 59.6, 76.4, 103.0, and 158.5 kev should be of particular interest in this study since according to the closed decay-energy cycle estimate an energy of about 0.22 Mev is available for the electron-capture decay of Pu^{237}.

II. EXPERIMENTAL TECHNIQUES

A. Sample Purification

The Pu^{237} was prepared in the Los Alamos Scientific Laboratory cyclotron by bombardments of enriched U^{235} foils with 2250 microampere hours of 26–29 Mev helium ions and 9280 microampere hours of 30-Mev helium ions. After bombardment, the foils were dissolved in dilute hydrochloric acid and the plutonium was purified by cycles of reduced and oxidized fluorides and elutions from anion resin columns with HI−HCl solution.[15] As a check on the isotopic purity, the decay of aliquots from each bombardment was followed for 5–7 months with a NaI(Tl) scintillation counter. Over this period only single-component decay was observed, and the half-life was found to be 45.6 days as previously reported[4] from this Laboratory.

B. 180° Permanent-Magnet Spectrograph

The conversion-electron lines of Pu^{237} were examined with a 119.5-gauss, 180° permanent-magnet spectrograph, and were recorded on Eastman no-screen medical

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[1] James, Florin, Hopkins, and Ghiorso, authors in *The Transuranium Elements: Research Papers*, edited by Seaborg, Katz, and Manning (McGraw-Hill Book Company, Inc., New York, 1949), National Nuclear Energy Series, Plutonium Project Record, Vol. 14B, Div. IV, Part II, p. 1604.

[2] James, Thompson, and Hopkins, authors in *The Transuranium Elements: Research Papers*, edited by Seaborg, Katz, and Manning (McGraw-Hill Book Company, Inc., New York, 1949), National Nuclear Energy Series, Plutonium Project Record, Vol. 14B, Div. IV, Part II, p. 1634.

[3] R. W. Hoff, University of California Radiation Laboratory Report UCRL-2325, 1953 (unpublished).

[4] D. C. Hoffman, J. Inorg. Nuclear Chem. **4**, 383 (1957).

[5] Thomas, Vandenbosch, Glass, and Seaborg, Phys. Rev. **106**, 1228 (1957).

[6] Jaffe, Passell, Browne, and Perlman, Phys. Rev. **97**, 142 (1955).

[7] F. Asaro and I. Perlman, Phys. Rev. **93**, 1423 (1954).

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

[9] S. A. Baranov and K. N. Shlyagin, *Proceedings Conference of the Academy of Sciences of the U.S.S.R. on the Peaceful Uses of Atomic Energy, Moscow, July 1–5, 1955*, Session of the Division of Physical and Mathematical Sciences (Akademiia Nauk, S.S.S.R., Moscow, 1955) [translation: Consultants Bureau, New York, 1955: U. S. Atomic Energy Commission Report TR-2436, 1956], Vol. 1, p. 183.

[10] Hollander, Smith, and Rasmussen, Phys. Rev. **102**, 1372 (1956).

[11] Wagner, Freedman, Engelkemeier, and Huizenga, Phys. Rev. **89**, 502 (1953).

[12] S. A. Baranov and K. N. Shlyagin, J. Exptl. Theoret. Phys. U.S.S.R. **30**, 225 (1956) [translation: Soviet Phys. JETP **3**, 200 (1956)].

[13] Bunker, Mize, and Starner, Bull. Am. Phys. Soc., Ser. II, **2**, 104 (1957).

[14] Rasmussen, Canavan, and Hollander, University of California Radiation Laboratory Report UCRL-3695, 1957 (unpublished).

[15] J. Kleinberg, Editor, Los Alamos Scientific Laboratory Report LA-1721, 1956 (unpublished).

x-ray film. The energy region from about 20 to 390 kev was observed with an energy resolution of ~0.2%. The Pu^{237} source was prepared by evaporation of the purified solution on a 0.010-in. diameter platinum wire which was then mounted in a standard aluminum holder in a reproducible position in the spectrograph. The conversion lines of Am^{241} sources[8,10] and Np^{239} sources[16] prepared in the same way were used in calibration. Day[8] has measured the absolute energies of the 26.36-, 33.20-, and 59.57-kev gamma rays from Am^{241} with an error of <0.1% with a curved-crystal diffraction spectrometer, and the conversion lines from Np^{239} have been measured with 180° permanent-magnet spectrographs by Hollander *et al.*[16] with an estimated absolute error of <0.3%. Therefore, in the energy region below 60 kev our absolute error is estimated to be ~0.2% where the lines are of reasonable intensity, while in the region above 60 kev it might be as much as 0.4%.

C. Beta-Ray Spectrometer

The beta-ray spectrometer employed in measuring the intensities of the conversion- and Auger-electron lines from the decay of Pu^{237} and Am^{241} was a uniform-field, ring-focusing solenoidal type patterned after the design of Schmidt.[17] The baffle system was set for nominal 2% transmission, and the measured resolution for the Pu^{237} and Am^{241} sources investigated was 0.8% and 1.0%, respectively. Although both sources were about 3/32 in. in diameter, and therefore would be expected to be observed with the same resolution, the slight residue in the Am^{241} source apparently was responsible for its being observed with significantly poorer resolution. These sources were prepared by evaporating small droplets of essentially carrier-free solutions of the two activities onto backings of aluminized-Mylar film (0.83 mg/cm²) and covering the deposits with thin Zapon films (5–10 μg/cm²).

The end-window counter of the spectrometer was a methane-flow, proportional counter operated at 10-cm Hg pressure with a loop-type anode. Two counter windows were used: from about 5 to 50 kev a thin Zapon window (~15 μg/cm²) supported by a 0.0005-in. thick Lektromesh grid was used; and from about 40 to 200 kev an aluminized-Mylar window (0.83 mg/cm²) was used. A study of the transmission characteristics of these windows was made by examining the Fermi-Kurie plot of the beta spectrum of a Pm^{147} source, as measured with each of these counter windows. The supported Zapon window was observed to have a constant transmission (57%) from 15 to 75 kev, and the transmission of the Mylar window was found to be constant (100%) above 45 kev.

[16] Hollander, Smith, and Mihelich, Phys. Rev. **102**, 740 (1956).
[17] F. H. Schmidt, Rev. Sci. Instr. **23**, 361 (1952).

D. Scintillation Spectrometer

The gamma-ray spectrum of Pu^{237} was examined with Harshaw-canned NaI(Tl) crystals mounted on DuMont 6292 photomultiplier tubes. Standard stabilized high-voltage supplies, nonoverload linear amplifiers, a coincidence circuit with a 0.35-μsec resolving time, and a fast 100-channel pulse-height analyzer were used with these scintillators. The Pu^{237} and Am^{241} sources used in these scintillation studies were prepared by evaporating small aliquots of the purified solutions on 1.75 mg/cm² Mylar film.

III. EXPERIMENTAL RESULTS

A. Gamma-Ray Spectra

The low-energy gamma-ray scintillation spectra of Pu^{237} and Am^{241}, taken under identical conditions, are shown in Fig. 1. The 26- and 60-kev photopeaks can be identified with the 26.4- and 59.6-kev gamma transitions which have been observed in the decay of Am^{241} and U^{237}. In the Pu^{237} spectrum, the Np *L* x-ray peak (~17 kev) is much more intense relative to the 26.4- and 59.6-kev photopeaks than in the Am^{241} spectrum. The most prominent feature of the Pu^{237} spectrum is the complex photopeak at ~100 kev which is attributed to Np *K* x-rays that arise as a result of *K*-electron capture. The gamma-ray spectrum of Pu^{237} above the Np *K* x-ray peak was also examined and an upper limit of 0.1% relative to the Np *K* x-rays was estimated for the intensity of any gamma rays above 120 kev. (This estimate was arrived at after weak peaks at ~160 and ~205 kev were shown to be the result of true-coincidence summing of the 60- and 100-kev pulses and chance coincidence "pileup" of the 100-kev pulses, respectively.)

The ratio of *K* x-radiation to *L* x-radiation to 59.6-kev radiation in Pu^{237} was determined by a direct comparison of the spectrum of Pu^{237} with that of Am^{241} at distances of 1, 7, and 11 cm, with and without a lead collimator. Corrections were made for escape-peak "losses" from the 60-kev and *K* x-ray peaks and for

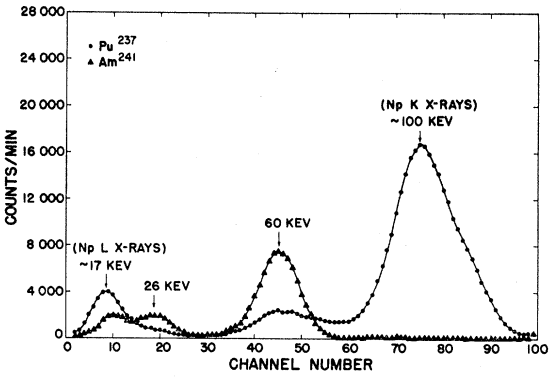

FIG. 1. Comparison of the gamma-ray spectra of Pu^{237} and Am^{241}, observed with a 1½-in.×1½-in. NaI(Tl) crystal at a distance of 1 cm from the sources.

absorption in the ~235-mg/cm² equivalent aluminum thickness of the can covering the NaI(Tl) crystal. (This absorber thickness was determined from a comparison of the experimentally determined ratio of the area of the 59.6-kev peak to the L x-ray peak in Am^{241} with the known photon intensity ratio of 0.97±0.06 reported by Beling *et al.*[18]) The following intensity ratios were then obtained for Pu^{237}: K x-radiation: L x-radiation: 59.6-kev gamma radiation = 1.00: 0.75 ±0.09: 0.14±0.02. Therefore, the number of 59.6-kev transitions per K-capture event is 0.30 if one uses 0.94 for the K-fluorescence yield (Sec. III D) and 1.3 for the total conversion coefficient of the 59.6-kev transition (calculated from the data of Barnov and Shlyagin[9] and Magnusson[19] for Am^{241}).

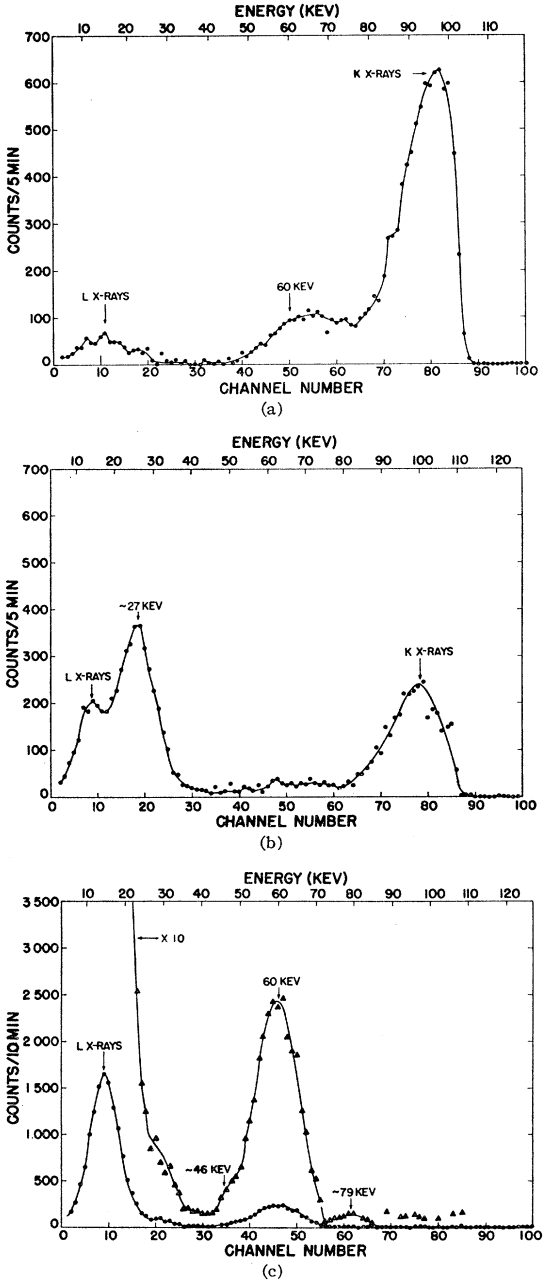

FIG. 2. Gamma-ray scintillation spectra of Pu^{237} in coincidence with gate pulses corresponding to the energy intervals: (a) 10–20 kev; (b) 51–67 kev; (c) 92–120 kev.

Coincidence measurements were made on a Pu^{237} sample placed 1 cm from each of two NaI(Tl) crystals ($1\frac{1}{2}$ in. thick × $1\frac{1}{2}$ in. diameter and 2 in. thick × 2 in. diameter) oriented at 180° to each other. The coincidence spectra obtained with the gate channel set to accept pulses corresponding to energy intervals of 10–20 kev (Np L x-ray region), 51–67 kev, and 92–120 kev (Np K x-ray region) are shown in Fig. 2. The chance coincidence background, obtained by putting a delay of 2.5 μsec in one of the coincidence channels, was measured and subtracted from the gross data in each case. Photons of ~17 (L x-rays), 59.6, and ~100 kev were found in coincidence with the L x-rays [Fig. 2(a)]. The coincidence spectrum gated by the 59.6-kev gamma rays [Fig. 2(b)] showed K and L x-ray peaks and a peak around 27 kev, while the spectrum in coincidence with the K x-ray region [Fig. 2(c)] showed the L x-ray peak and the 59.6-kev photopeak. A low-intensity peak is indicated at 79 kev which is predominantly the result of true-coincidence summing of L x-radiation and the 59.6-kev photons.

The ~27-kev peak shown in Fig. 2(b) was not present in the coincidence spectrum taken with the scintillators at 60° to each other with lead absorber between them. Therefore, this peak can be attributed to scattering processes or the iodine K x-rays which are in coincidence with the escape peak pulses from the ~100-kev photons. The other measurements were also repeated at 60°, but no significant change in the spectra could be detected.

The fact that K x-rays are in coincidence with 59.6-kev photons establishes the existence of K capture to the 59.6-kev level of Np^{237} and sets a minimum energy of ~180 kev (59.6 kev + K-binding energy of 118.6 kev) for the electron-capture decay of Pu^{237}. The ratio of L to K x-radiation in coincidence with the 59.6-kev gamma ray [Fig. 2(b)] was found to be 1.9, compared with the corresponding ratio of 0.75 found in the ungated spectrum. This ratio is an upper limit since there is a small contribution to the gated L x-ray peak from L capture to the 103.0- and 158.5-kev levels, and from coincidences due to the inclusion of some of the K x-ray escape peak in the 51- to 67-kev gate interval.

B. Electron Spectra

The electron data obtained for Pu^{237} from the permanent-magnet spectrograph are summarized in Table I. In addition to the well known 33.20-, 43.46-, and 59.57-

[18] Beling, Newton, and Rose, Phys. Rev. **86**, 797 (1952); **87**, 1144 (1952).

[19] L. B. Magnusson, Phys. Rev. **107**, 161 (1957).

TABLE I. Conversion electron lines from permanent-magnet spectrograph.

Electron energy (kev)	Shell	Transition energy[a] (kev)	Visual intensity estimate[b]
		(26.36)[c]	not seen
		(33.20)[c]	
27.53	M_{I}	33.27	m
27.81	M_{II}	33.17	w
28.79	M_{III}	33.22	vw
29.43	$M_{IV,V}$	33.18	vvw
31.80 complex	$N_{I,II,III}$	~33.2	wm
32.35 31.68–31.98	$N_{IV,V}$	33.15	vw
32.84	O	~33.2	vw
		(43.46)[c]	
21.09	L_{I}	43.51	vvw
21.89	L_{II}	43.49	vvw
25.97	L_{III}	43.58	vvvw
37.84	M_{I}	43.58	vvvw
39.09	M_{III}	43.52	vvw
~42.8	N	~43.9	vvw
~43.4	O	~43.7	vvw
		(55.56)[c]	
33.08	L_{I}	55.50	vvw
33.83	L_{II}	55.43	vvw
		(59.57)[c]	
37.19	L_{I}	59.61	m
38.06	L_{II}	59.66	ms
41.99	L_{III}	59.60	m
53.80	M_{I}	59.54	m
54.25	M_{II}	59.59	m
55.18	M_{III}	59.61	w
55.8	$M_{IV,V}$	~59.5	vvw
58.06	N_{I}	59.56	wm
58.30	N_{II}	59.62	wm
58.53	N_{III}	59.61	vw
59.33	$O_{I,II}$	~59.6	w
		76.4[d]	
54.78	L_{II}	76.38	w
70.56	M_{I}?	76.30	vw
71.16	M_{II}	76.52	vw
72.1	M_{III}	76.5	vvw
73.0	$M_{IV,V}$	~76.7	vvw

[a] Binding energies from tables given by Hill, Church, and Mihelich [Rev. Sci. Instr. **23**, 523 (1952)], and S. Fine and C. F. Hendee [Nucleonics **13**, No. 3, 36 (1955)].
[b] s=strong; w=weak; m=moderate; v=very.
[c] Transition energies in parentheses are from references 8 and 10.
[d] This energy value was determined from the L_{II}- and M_{II}-conversion lines.

kev transitions which have been observed in the decay of Am^{241} [6–10] and U^{237},[11–14] evidence for a 76.4-kev gamma ray has been found. This may be the transition from the 76.1-kev rotational level, 9/2+, found by Newton[20] in the Coulomb excitation of Np^{237}. It has not been observed in the decay of Am^{241} or U^{237}. The transition should be pure $E2$ and hence we would not expect to see the L_{I}- and M_{I}-conversion electron lines on our film spectrogram. However, a line was seen at 70.56 kev which would correspond to the M_{I} line of a ~76-kev transition. The presence of the L_{I} line (~54 kev) could not be established because of interference from the relatively strong M_{I} (53.80 kev) and M_{II} (54.25 kev) lines from the 59.57-kev transition. Furthermore, for an $E2$ transition, the L_{III} line should be comparable in intensity to the observed intensity of the L_{II} line, but the L_{III} line would be masked by the N and O lines arising from conversion of the 59.57-kev gamma ray. A "very, very weak" line at 72.1 kev might be the M_{III} line of the 76.4-kev transition, although it should be comparable in intensity to the M_{II} line. Despite the fact that the relative intensities of the conversion lines of the 76.4-kev transition are not in complete agreement with the expected $E2$ character, it has been tentatively assigned as the ground-state transition from the 9/2+ rotational level.

There is some evidence for extremely weak L_{I} and L_{II} lines from the 55.56-kev transition which has been seen in the alpha decay of Am^{241}. Its presence would establish the existence of electron capture to the known 158.5-kev level of Np^{237} and confirm a minimum disintegration energy of ~180 kev if one assumes L capture to this level.

Figure 3 shows the electron spectra of Pu^{237} and Am^{241} from ~3 kev to 60 kev obtained with the beta-ray spectrometer employing the Zapon-window counter. No correction was applied to either spectrum below 15 kev, where window absorption cuts down the intensity. The electron line assignments and the relative intensities of those lines which could be measured with some certainty are summarized in Table II.

TABLE II. Summary of conversion-electron line data from beta-ray spectrometer measurements on Am^{241} and Pu^{237} (Figs. 3 and 4).

Line No.	Designation[a]	Line energy (kev)	Reported energy[b] (kev)	Relative intensity[c] Pu^{237}	Relative intensity[c] Am^{241}
1	L_{III}(26.4)Am	8.7	8.75[d]		
2	L_{I}(33.2)Am	10.8	10.82		
3	L_{II}(33.2)Am	11.7	11.62		
4	L_{III}(33.2)Am	15.2	15.55		
5	L-Augers Pu	19.6–20.4			
6	M_{I}(26.4)Am, L_{I}(43.5)Am	20.9	20.61, 21.05		
7	L_{II}(43.5)Am	21.6	21.87		
8	N_{I}(26.4)Am	24.9	24.84[d]		
9	L_{III}(43.5)Am	25.8	25.85		
10	M_{I}(33.2)Pu, M_{II}(33.2)Pu	27.4	27.46, 27.84	244	27
11	M_{III}(33.2)Pu	~28.6	28.80		
12	N_{I}(33.2)Pu, N_{II}(33.2)Pu	31.5	31.73, 31.92	121	...
13	O_{I}(33.2)Pu, O_{II}(33.2)Pu	32.8	32.87		
14	L_{II}(55.6)Am	34.0	33.99		
15	L_{I}(59.6)Pu, Am	37.1	37.16		
16	M_{I}(43.5)Pu, Am; L_{II}(59.6)Pu, Am	37.9	37.71, 37.98	337	339
17	M_{III}(43.5)Am	~39.0	39.05		
18	L_{III}(59.6)Pu, Am	41.9	41.96	59	59
19	M_{I}(59.6)Pu, Am; M_{II}(59.6)Pu, Am	54.0	53.83, 54.21	100	100
20	N_{I}(59.6)Pu, Am; N_{II}(59.6)Pu, Am	58.1	58.07, 58.25	34.4	36
21	M(76.4)Pu	...	...	<10	

[a] Designation refers to the electron vacancy, the transition energy in kev, and the spectrum (Am^{241} or Pu^{237}) from which the line energy was read.
[b] These values are taken from the film spectrographic measurements of Hollander, Smith, and Rasmussen, (reference 10).
[c] Line intensities were normalized to th $M_{I,II}$ (59.6 kev) line in each spectrum.
[d] These two energy values are from the work of Baranov and Shlyagin (reference 9).

[20] J. O. Newton, Harwell Meeting of the Physical Society, Nature **175**, 1027 (1955); see p. 1028.

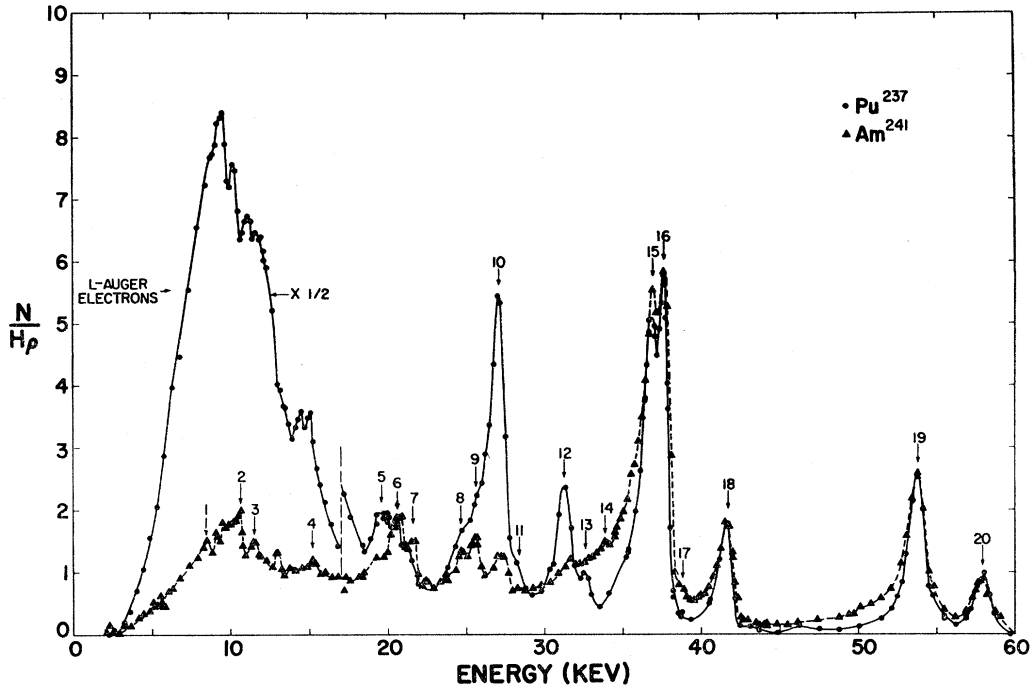

FIG. 3. Conversion-electron and L-Auger electron spectra of Pu^{237} and Am^{241}, obtained with the beta-ray spectrometer using the Zapon-window counter.

C. K-Auger Electron Lines

The K-Auger data from the permanent-magnet spectrograph and the beta-ray spectrometer are summarized in Table III. Figure 4 shows the Pu^{237} K-Auger electron spectrum obtained with the beta-ray spectrometer equipped with the Mylar-window counter. The $M_{I,II}$ (59.6)- and $N_{I,II}$ (59.6)-conversion electron lines are included for intensity comparison. A careful search from 112 kev (line K_{10}) up to 190 kev revealed no detectable electron lines.

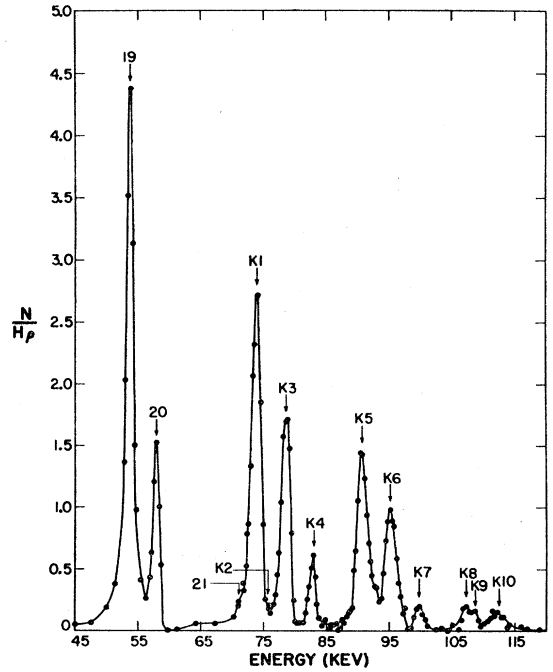

FIG. 4. K-Auger electron spectrum of Pu^{237} obtained with the beta-ray spectrometer equipped with the Mylar-window counter.

The film spectrograph lines at 73.7 and 74.6 kev appear to be quite broad for single lines and may be 73.6-, 73.8-kev and 74.5-, 74.8-kev doublets, respectively. A questionable weak line was also observed at 78.15 kev. However, the film was very difficult to read in these regions because of background darkening resulting from the long exposure time required for the relatively weak Pu^{237} source. Even for the K-$L_{II}L_{III}$ line, which is quite sharp, the energy of 79.32 kev from the film spectrograph seems to be too high since the calculated energy is 79.0 kev. The film spectrograph energies for the other K-LL vacancies also appear to be $\sim$0.3 kev higher than the calculated values. The discrepancy might be explained by the uncertainty in the K-shell binding energy for $Z=93$, but Hollander *et al.*[16] have experimentally confirmed the value of 121.8 kev computed[21,22] for $Z=94$ by extrapolation from values given by Cauchois,[23] and we have used the corresponding value of 118.6 for $Z=93$. However, since the error in our absolute energy calibration could be as much as 0.4% (see Sec. II B) there may be no real energy discrepancy. The K-LX energies (where X denotes the M, N, etc., orbital-

[21] Hill, Church, and Mihelich, Rev. Sci. Instr. **23**, 523 (1952).
[22] S. Fine and C. F. Hendee, Nucleonics **13**. No. 3, 36 (1955).
[23] Y. Cauchois, J. phys. radium **13**, 113 (1952).

TABLE III. K-Auger lines from Pu^{237} decay.

Line No.	Energy (kev) β-ray spectrometer	Energy (kev) Film spectrograph	Transition	Calculated energy[a] (kev)	Relative intensity Film spectrograph	Relative intensity β-ray spectrometer[b]
K_1	73.9	73.7[c]: 73.6, 73.8	K-L_IL_I	73.4	w, m	74.6
		74.6[c]: 74.5, 74.8	K-L_IL_{II}	74.2	m, m	
K_2	not resolved	75.3	K-$L_{II}L_{II}$	75.0	vvw	≦5.5
K_3	78.6	78.15	...		w	44.3
		78.52	K-L_IL_{III}	78.2	wm	
		79.32	K-$L_{II}L_{III}$	79.0	wm	
K_4	82.9	83.3	K-$L_{III}L_{III}$	83.0	w	11.4
K_5	90.6	90.74	K-$L_IM_{I,II,III}$	90.5, 90.8, 91.8	w	41.6
		91.22	K-$L_{II}M_{I,II,III}$	91.3, 91.6, 92.6	w	
K_6	95.2	94.95	K-$L_IN_{I,II,III}$	94.6, 94.9, 95.1	vw	32.6
		95.43	K-$L_{III}M_{I,II}$ or K-$L_{II}N_{I,II}$	95.3, 95.6 / 95.4, 95.7	vw	
		96.11	K-$L_{II}N_{III}$ or K-$L_{III}M_{III}$	95.9 / 96.6	vw	
K_7	99.8	not seen	K-$L_{III}N_{I,II,III}$	99.50–99.92	...	5.1
K_8	107.3	not seen	K-$M_{I,II}M_{I,II}$	107.1–107.9	...	3.4
K_9	108.7	not seen	K-$M_{I,II}M_{III}$	108.4–108.8	...	2.8
K_{10}	112.0	not seen	K-$M_{I,II}N_{I,II,III}$	111.4–112.4	...	5.4

[a] I. Bergström and R. D. Hill, Arkiv Fysik **8**, 21 (1954).
[b] Intensities normalized to $M_{I,II}$ (59.6) line, No. 19 (Mylar-window counter) =100. The accuracy of the relative intensities is estimated to be about 5%.
[c] These lines appeared fuzzy and broader than the usual line and may actually be composed of the two lines indicated.

electron shells) appear to be in agreement with the calculated values, although this is difficult to determine since most of these lines are complex.

If the higher energy components of the possible doublets at 73.7 and 74.6 kev are assigned to the K-L_IL_I and K-L_IL_{II} Auger transitions, then we have unassigned lines at 73.6, 74.5, and 78.15 kev. These could correspond to the L_I-, L_{II}-, and L_{III}-conversion lines of a 96.0-kev gamma ray. A stronger source of Pu^{237} will have to be examined in order to prove or disprove this interpretation of these lines. In the rotational band which includes the 33.2-kev level and the new level at 76.4 kev, the next level $(11/2+)$ would be at ~128 kev. There would then be the possibility of an $E2$ transition of ~95 kev between the 128- and 33.2-kev levels if the 128-kev level is populated either directly by electron capture or from the 158.5-kev level. Direct capture to the 128-kev level would be relatively small since the spin of Pu^{237} is probably $\frac{7}{2}$ (see Sec. V). If the level is fed by a transition from the 158.5-kev level it should have been observed in the decay of Am^{241}. Therefore, such placement of a 96-kev transition seems rather improbable.

Values of 0.58±0.03 for the ratio of L-LX/K-LL Auger electrons, and 0.085±0.008 for the ratio of K-XX/K-LL Auger electrons for $Z=93$ have been calculated by summing the line intensities obtained from the beta-ray spectrometer (Table III). The value of 0.58 appears to be consistent with experimental values of this ratio tabulated by Gray[24] for other elements. A comparison of the relative intensities of individual K-LL Auger electron lines is difficult since the beta-ray spectrometer did not resolve all of these, and only qualitative intensities were obtained from the film spectrograms. However, a comparison of the ratio of

TABLE IV. Relative intensities of K-LL Auger lines.

Element	Z	$(K\text{-}L_IL_{III})$ / $+(K\text{-}L_{II}L_{III})$	K-$L_{III}L_{III}$	K-$L_{III}L_{II}$	$(K\text{-}L_IL_I)$ / $+(K\text{-}L_IL_{II})$	Reference
silver	47	1.0	0.40	0.11	0.51	a
gold	79	1.0	0.31	0.12	1.04	b
mercury	80	1.0	0.29	0.10	1.05	c
bismuth	83	1.0	0.30	<0.07	1.04	d
bismuth	83	1.0	0.36	<0.06	0.78	e
polonium	84	1.0	0.37	<0.05	1.26	f
neptunium	93	1.0	0.26	<0.12	1.68	g
plutonium	94	1.0	0.27	0.09	1.21	h
theoretical nonrelativistic		1.0	0.37	0.06	0.34	i

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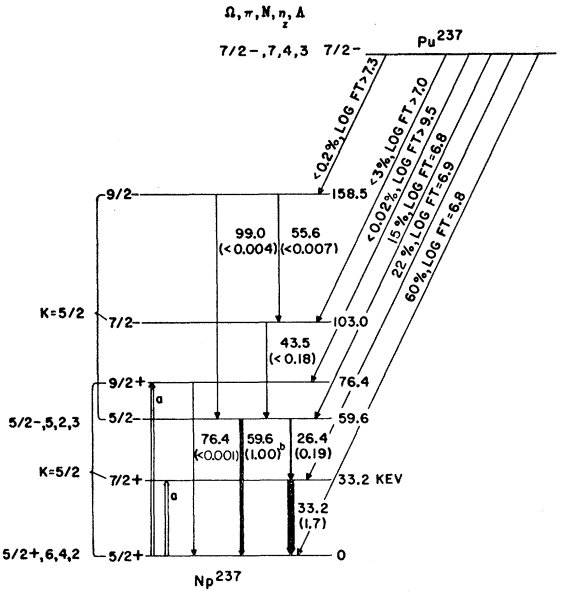


FIG. 5. Proposed electron-capture decay scheme of Pu^{237}. The spins, parities, and asymptotic quantum numbers assigned to the levels of Np^{237} are those proposed by Hollander *et al.*[10] and Rasussen *et al.*[14] (a) Levels observed in Coulomb excitation experiments.[20] (b) The values in parentheses give estimates of the intensities of the various transitions relative to the 59.6-kev transition.

$(K\text{-}L_IL_I+K\text{-}L_IL_{II}):(K\text{-}L_{II}L_{II}):(K\text{-}L_{III}L_{III}):(K\text{-}L_IL_{III}+K\text{-}L_{II}L_{III})$ with other experimental results and with the theoretical nonrelativistic calculations has been made in Table IV. Although our relative intensities for neptunium are not at all in agreement with the theoretical predictions, they seem to be in fair agreement with the experimental values for other heavy elements. However, it appears that the intensity of the $(K\text{-}L_IL_I+K\text{-}L_IL_{II})$ Auger electrons is too high, which again might indicate that there is some contribution from the L-conversion lines of a 96.0-kev transition.

The K-Auger electron coefficient was calculated from a comparison of the K x-ray intensity as observed with the scintillation spectrometer and the K-Auger electron intensities measured with the beta-ray spectrometer. The intensity ratio of K-Auger electrons to the sum of M_I- and M_{II}-conversion electrons (arbitrary intensity = 100) from the 59.6-kev transition in the decay of Pu^{237} was found to be 2.26 by summing the intensities of all of the K-Auger lines listed in Table III. The data of Baranov and Shlyagin[9] and Magnusson[19] for the ratios of the intensities of these conversion electrons and the ratio of the intensity of 59.6-kev gamma rays to alpha distintegrations in Am^{241} of 0.36 were used to calculate a value of 0.21 for the M_I- and M_{II}-conversion coefficient of the 59.6-kev transition. The intensity ratio of K-Auger electrons to 59.6-kev gamma rays was then calculated to be 0.47. From the ratio of 0.14 for 59.6-kev gamma rays to K x-radiation obtained from the scintillation data, the ratio of K-Auger electrons per K-shell vacancy, or the K-Auger coefficient, was calculated to be 0.062 ± 0.01. The corresponding K-fluorescence yield, $\bar{\omega}_K$, is then 0.94 for $Z=93$. This is somewhat lower than the value of 0.97 calculated from Gray's fit[24] of the expression, $\bar{\omega}_K=AZ^4(b+Z^4)^{-1}$, to the experimental K-fluorescence yields of the heavier elements.

IV. ENERGY LEVELS OF Np^{237} POPULATED BY ELECTRON-CAPTURE DECAY OF Pu^{237}

The evidence for the existence of 26.36-, 33.20-, 43.46-, 59.57-, 55.56-, and 76.4-kev gamma transitions accompanying the decay of Pu^{237} has been described in the preceding sections. These data indicate that the previously established 33.20-, 59.57-, 103.0-, and 158.5-kev levels of Np^{237} are fed by the electron-capture decay of Pu^{237}. The postulated 76.4-kev level $(9/2+)$ appears to be populated principally by direct electron-capture since there is no evidence that it is fed by transitions from upper levels of Np^{237}. From a comparison of the electron spectra of Pu^{237} and Am^{241}, the intensities of the various transitions relative to the 59.6-kev transition in Pu^{237} were estimated (Fig. 5).

It is obvious from Fig. 3, line 10, that the intensity of the 33.2-kev transition is much greater in the Pu^{237} decay than in the Am^{241} decay. It was calculated to be 9.0 times (244/27 from Table II) as intense by comparing the relative intensities of the M_I- and M_{II}- conversion lines of the 33.2-kev transition. Values of 84% and 16% were calculated for the de-excitation of the 59.6-kev state by the 59.6-kev transition and the 26.4-, 33.2-kev cascade, respectively. (The following data were used in the calculation: the ratio of all of the conversion lines of the 59.6-kev transition relative to those of the 33.2-kev transition in Am^{241} is 0.34;[9] the number of 59.6-kev photons per Am^{241} alpha disintegration is 0.36;[19] 99.4% of the Am^{241} alpha disintegrations populate[6,7] the 59.6-kev level and only 0.2% directly feed the 33.2-kev level; and the 33.2-kev transition is almost completely converted.) Therefore, in the decay of Pu^{237}, the ratio of the 33.2- to the 59.6-kev transition was calculated to be 1.7.

The ratio of the 26.4-kev to 59.6-kev gamma transition must, of course, be the same for Pu^{237} as for Am^{241} and is 0.19. It was estimated (see Fig. 3, lines 6, 7, and 17) that the 43.4-kev transition in Pu^{237} is weaker relative to the 59.6-kev transition than it is in the Am^{241} spectrum. The L_{II}-conversion line from the 55.6-kev transition was seen in the Am^{241} spectrum (Fig. 3, line 14) but was not resolved in the Pu^{237} spectrum, which indicates that it is probably less intense in the decay of Pu^{237}. However, a visual comparison of the intensities of the L_I- and L_{II}-conversion lines from film spectrograms of Pu^{237} and Am^{241} indicated that the intensity of the 55.6-kev transition relative to that of the 59.6-kev transition was roughly comparable in the two decays. From alpha spectroscopic data,[6,7] it has been determined that $\sim$15% of the total alpha decay of Am^{241} goes through the 103.0-kev level, which is

depopulated by the 43.4-kev transition. Therefore, the 43.4-kev to 59.6-kev transition intensity ratio in the decay of Am^{241} is ~ 0.18 and it is somewhat less than this in the decay of Pu^{237}. A similar estimate for the 55.6-kev transition of Pu^{237} indicates that its intensity relative to that of the 59.6-kev transition is $<0.7\%$.

No evidence could be found for the known 99.0-kev transition which is one mode of de-excitation of the 158.5-kev level of Np^{237}. However, the L_{I}- and L_{II}-conversion lines would be masked by the K-Auger lines on the beta-ray spectrometer plots. Since these lines should be even less intense than those from the 55.6-kev transition which also depopulates the 158.5-kev level and whose L_{I}- and L_{II}-conversion lines could barely be detected, one would not expect to be able to see any conversion lines from the 99.0-kev transition on the film spectrograms.

It was not possible completely to resolve any conversion lines of the 76.4-kev transition in the beta-ray spectrometer plots. However, from the small contribution to the line spectrum in the 70–72-kev region (line 21, Fig. 4), an upper limit of 10% was estimated for the intensity of the M lines of this transition relative to the intensity of the M lines of the 59.6-kev transition. By means of a computed M-conversion coefficient[25] of about 20 for a 76-kev $E2$ transition and an M-conversion coefficient[9] of 0.25 for the 59.6-kev transition, an upper limit of 0.13% was placed on the intensity of the 76.4-kev transition relative to the 59.6-kev transition.

V. CALCULATIONS AND DISCUSSION

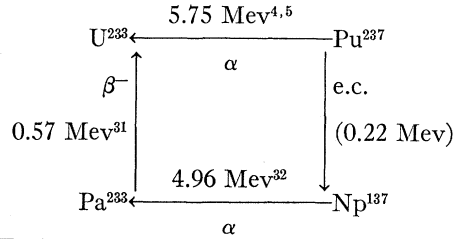
The energy available for electron-capture decay can be calculated[26] from the ratio of L_{I}- to K-electron capture for allowed or first-forbidden transitions ($\Delta I=0,1$)[27] if the energy is near the K-capture threshold. This ratio was calculated for electron-capture decay to the 59.6-kev level in Np^{237} from the experimental value of 1.9 for the relative intensity of L to K x-rays in coincidence with 59.6-kev gamma rays. Corrections for the L x-radiation arising from processes other than L-electron capture, such as L x-rays following K-electron capture and L-conversion processes, were made as shown in Eq. (1):

$$\frac{L_{\mathrm{capt}}}{K_{\mathrm{capt}}}=\frac{L_{\mathrm{x}}}{K_{\mathrm{x}}}\left(\frac{\bar{\omega}_K}{\bar{\omega}_L}\right)-\frac{L_K}{K_{\mathrm{capt}}}-\bar{\omega}_K\left(\frac{L_{\mathrm{conv}}}{K_{\mathrm{x}}}\right), \qquad (1)$$

where $L_{\mathrm{x}}/K_{\mathrm{x}}$ is the observed L to K x-ray intensity ratio; $\bar{\omega}_K$ and $\bar{\omega}_L$, the K- and L-fluorescence yields, are taken to be 0.94 and 0.5,[28,29] respectively; L_K/K_{capt}, the fraction of K-shell vacancies filled by L-shell electrons, is 0.72[30]; and L_{conv} is the number of L vacancies arising from L conversion of the appropriate gamma transitions. For electron capture to the 59.6-kev level, substitution in Eq. (1) gives:

$$\left(\frac{L_{\mathrm{capt}}}{K_{\mathrm{capt}}}\right)_{59.6}=\left[1.9\left(\frac{0.94}{0.5}\right)-0.72-0.97\left(\frac{\Sigma e^- L_{56,43}}{\gamma_{60}}\right)\left(\frac{\gamma_{60}}{K_{\mathrm{x}}}\right)\right]=2.8\pm 0.8. \qquad (1a)$$

The ratio, $(\Sigma e^- L_{56,43}/\gamma_{60})$, was calculated to be 0.3 from the intensities of the L-conversion electron lines of the 55.6-kev and 43.4-kev transitions relative to those of the 59.6-kev transitions using a value[9,19] of 0.97 for the L-conversion coefficient of the 59.6-kev transitions. The number of 59.6-kev gamma rays per K x-ray, $(\gamma_{60}/K_{\mathrm{x}})$, is 0.14 as determined from the ungated gamma spectrum of Pu^{237}. The corrections for L x-rays from K capture to the 59.6-kev level and L x-rays from the L conversion of the relatively weak 43.4- and 55.6-kev transitions, which are in coincidence with the 60-kev photons, are quite small. Therefore, the ratio computed from Eq. (1a) is affected very little by the values used for the fraction of K-shell vacancies filled by L-shell electrons and the L-conversion coefficient of the 59.6-kev transition. According to Brysk and Rose,[26] the L_{III}- to L_{I}-capture ratio is negligibly small for allowed and first-forbidden transitions, whereas the L_{II}- to L_{I}-capture ratio is ~ 0.13 for $Z=94$, and is quite insensitive to the order of forbiddenness. The ratio of L- to K-electron capture, after this correction for L_{II}-electron capture, yields an energy of 153 kev for the transition to the 59.6-kev level. The total electron-capture decay energy is then 0.21 Mev which is in satisfactory agreement with the electron-capture decay energy of 0.22 Mev estimated from the closed decay-energy cycle shown below.[31,32]



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[28] Strictly, a different $\bar{\omega}_L$ is associated with each of the various processes resulting in L x-radiation since the fluorescence yield is different for each L shell and the probability for ionization of a given shell is different for L-electron capture, the conversion of gamma rays, and the L vacancies arising after K capture. However, a mean value[29] of 0.5 was used for $\bar{\omega}_L$.

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The intensity of electron capture to the ground state of Np^{237} relative to the 59.6-kev transition was calculated[33] from the intensities of the 33.2-, 59.6-, and 76.4-kev gamma transitions relative to the 59.6-kev transition (Fig. 5) using the experimental value of 0.30 for the number of 59.6-kev transitions per K-capture event and the theoretical L- to K-capture ratios[26] for capture to the various levels. The estimates of the relative intensities of the various transitions and the resulting branching ratios are shown in Fig. 5, the proposed electron-capture decay scheme of Pu^{237}. The logft values were calculated using a total electron-capture decay energy of 0.21 Mev. The spins, parities, and asymptotic quantum numbers, Ω, N, n_z, and Λ, are those assigned by Hollander *et al.*[10] and Rasmussen *et al.*[14] Our data indicate that the electron-capture decay of Pu^{237} is predominantly to the $\frac{5}{2}+$, $\frac{7}{2}-$, and $\frac{5}{2}-$ levels of Np^{237} with corresponding logft values of 6.8, 6.9, and 6.8, respectively, which are consistent with the logft values tabulated by Alaga[34] for first forbidden ($\Delta I=0, 1$) or hindered allowed transitions. Therefore, one would postulate a $\frac{5}{2}$ or $\frac{7}{2}$ spin for the ground state of Pu^{237}. U^{235}, with the same number of neutrons but with two fewer protons than Pu^{237}, has a spin[35] of $\frac{7}{2}$ and a measured magnetic moment[36] of -0.3. Inspection of the portion of the Nilsson energy level diagram presented by Hollander[37] shows that states of $\frac{7}{2}-(7,4,3)$ lying close to the $\frac{1}{2}-(6,3,1)$ level postulated[37] for Pu^{239} and $\frac{7}{2}+(6,2,4)$ close to the $\frac{1}{2}-(5,0,1)$ state proposed[14] for U^{237}, are available in this region. [The Nilsson notation for a level is $\Omega\pi(N,n_z,\Lambda)$, where Ω is the spin, π the parity, N the total oscillator quantum number, n_z the component of N along the symmetry axis, and Λ the component of the particle orbital angular momentum l along the symmetry axis.] However, since the theoretical values of the magnetic moment, calculated according to the method of Nilsson,[38] are ~-0.6 for the $\frac{7}{2}-(7,4,3)$ orbit and $\sim+1.1$ for the $\frac{7}{2}+(6,2,4)$ orbit, the $\frac{7}{2}-(7,4,3)$ orbit is assigned to the U^{235} ground state. By analogy, we have also assigned the $\frac{7}{2}-(7,4,3)$ orbit to the Pu^{237} ground state although a neighboring $\frac{5}{2}+(6,3,3)$ state cannot be definitely ruled out on the basis of the present experimental data.

ACKNOWLEDGMENTS

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[33] The calculation was made from the following relationship: $K_{\text{capt}}/I_{59.6}=\Sigma_i(K_i/I_{59.6})$, where $K_i/I_{59.6}$ is the intensity of K capture to the ith level relative to the intensity of the 59.6-kev transition and i denotes the 0-, 33.2-, 59.6-, or 76.4-kev state. (The total decay energy of 0.21 Mev is insufficient to allow K capture to the higher energy levels.) Since the total number of K- and L-capture events to a given level must be equal to the intensity of the transitions de-exciting the level (corrected for the contribution of transitions populating it from upper levels), $\Sigma_i(K_i/I_{59.6})=\Sigma_i(I_i/I_{59.6})(1+L_i/K_i)^{-1}$, where I_i is the corrected intensity of the transitions de-exciting the ith level and L_i/K_i is the theoretical L- to K-capture ratio[26] for electron-capture decay to the ith level. Then $I_0/I_{59.6}=K_{\text{capt}}/I_{59.6}-(I_j/I_{59.6})(1+L_jK_j)^{-1}$, where j refers to the 33.2-, 59.6-, or 76.4-kev level. Proper substitution in the equation gives $I_0/I_{59.6}=4.1$ or 60% of the electron capture is to the ground state.

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