Radiations of 6.7-Day U^{237}

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(Received September 19, 1952)

The radiations of U^{237} have been studied using a magnetic lens beta-ray spectrometer and scintillation spectrometers for betas and gammas singly and in coincidence. A beta with an end-point energy of 245±5 kev was found along with evidence for the existence of a lower energy component. Gammas of energy 27, 43, 59, 165, 207, 269, 334, 370, and 430 kev were found, and probably a gamma of 102 kev. The decay scheme constructed gives a total decay energy for this isotope of 551±5 kev.

INTRODUCTION

SINCE U^{237} was first observed by Nishina *et al.*[1] and independently by McMillan,[2] several studies[2–5] of the radiations from this isotope have been made, but none have led to a definite assignment of the total decay energy, and there has been a lack of agreement among the several investigators as to the character of these radiations.

McMillan's[2] absorption data indicated the presence of a simple beta-ray ($E_{max}=0.25$ Mev) and two soft gamma-rays. Later work by Brady and Rubinson,[3] counting with the use of aluminum and lead absorbers, led them to assign the activity to three beta-particles ($E_{max}=1.5$, 1.2, and 0.25 Mev) and to gammas of energies 0.53, 0.23, and 0.14 Mev.

A more extensive study of this isotope by Engelkemeir and Turkevich[4] using coincidence methods as well as straight absorption counting enabled them to give a more detailed picture of these radiations. They reported gammas of 0.050, 0.220, and 0.450 Mev, conversion electrons at *ca* 0.1 and *ca* 0.3 Mev, and a beta-continuum with $E_{max}=0.3$ Mev. They also detected the presence of neptunium x-rays (14 and 95 kev).

More recently Melander and Slätis[5] studied the beta-ray and conversion line spectrum of samples of U^{237} with a magnetic lens spectrometer. They reported the presence of a beta-continuum ($E_{max}=0.23$ Mev), possibly a lower energy beta, and eight conversion lines corresponding to gammas of energy 0.057, 0.204, and 0.260 Mev. Gamma-absorption measurements were in agreement with these results.

The experiments reported here were undertaken in order to furnish data as to the total decay energy of the U^{237} nucleus for use in formulating the energy cycles among the heavy elements.[6]

EXPERIMENTAL

The U^{237} used in these experiments was formed by neutron irradiation of enriched uranium (*ca* 50 percent U^{236}) in the Argonne heavy water pile.

After irradiation uranium was chemically separated from fission products and other activities by the following chemical procedure.

1. Uranyl nitrate was dissolved in HNO_3 after irradiation.
2. $(NH_4)_2U_2O_7$ was precipitated; the precipitate was then washed.
3. $(NH_4)_2U_2O_7$ was dissolved in concentrated HNO_3. Urea and Fe^{++} were added (to reduce neptunium to Np^{+4}), the solution was diluted to 0.4*M* H^+ and saturated with NH_4NO_3.
4. Uranium was diethyl ether extracted from aqueous solution, washed with saturated NH_4NO_3, and then re-extracted back into water (activities such as iodine remain in ether).
5. The water solution was re-extracted with ether several times to remove traces of halogens.
6. $NaUO_2(Ac)_3$ was precipitated from the water solution.
7. Steps 2 to 6 were repeated five times, omitting step 6 in the last cycle.

The final extraction resulted in a solution of uranyl nitrate in water.

Evidence for the decontamination achieved with this procedure is the fact that no change was observed in the spectrum taken on a sample after several additional extraction cycles were performed. A sample showed a linear decay over a period of thirteen half-lives in agreement with the 6.75±0.01 days half-life measured by Huizenga and Flynn.[7]

BETA-RAY SPECTROMETER STUDIES

The samples for the beta-ray spectrometer were prepared by evaporating a solution of uranium on thin (10 $\mu g/cm^2$) LC-600 films, which were made conducting by a coat of Aquadag on the back side.

The instrument used was a double lens magnetic spectrometer having a transmission of *ca* 2 percent and a resolution for the Ba^{137} conversion line of *ca* 2 percent.

The detector was an atmospheric pressure flow-type methane proportional counter. For energies below 135

[1] Nishina, Yaseki, Ezoe, Kimura, and Ikawa, Phys. Rev. **57**, 1182 (1940).
[2] E. McMillan, Phys. Rev. **58**, 178 (1940).
[3] E. L. Brady and W. Rubinson, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), Paper No. 3.21, National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV.
[4] D. W. Engelkemeir and A. Turkevich, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), Paper No. 3.22, National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV.
[5] L. Melander and H. Slätis, Arkiv Mat. Astron. Fysik **A36**, No. 15 (1948–1949).
[6] Huizenga, Magnusson, Freedman, and Wagner, Phys. Rev. **84**, 1264 (1951).
[7] J. R. Huizenga and K. F. Flynn (unpublished results).

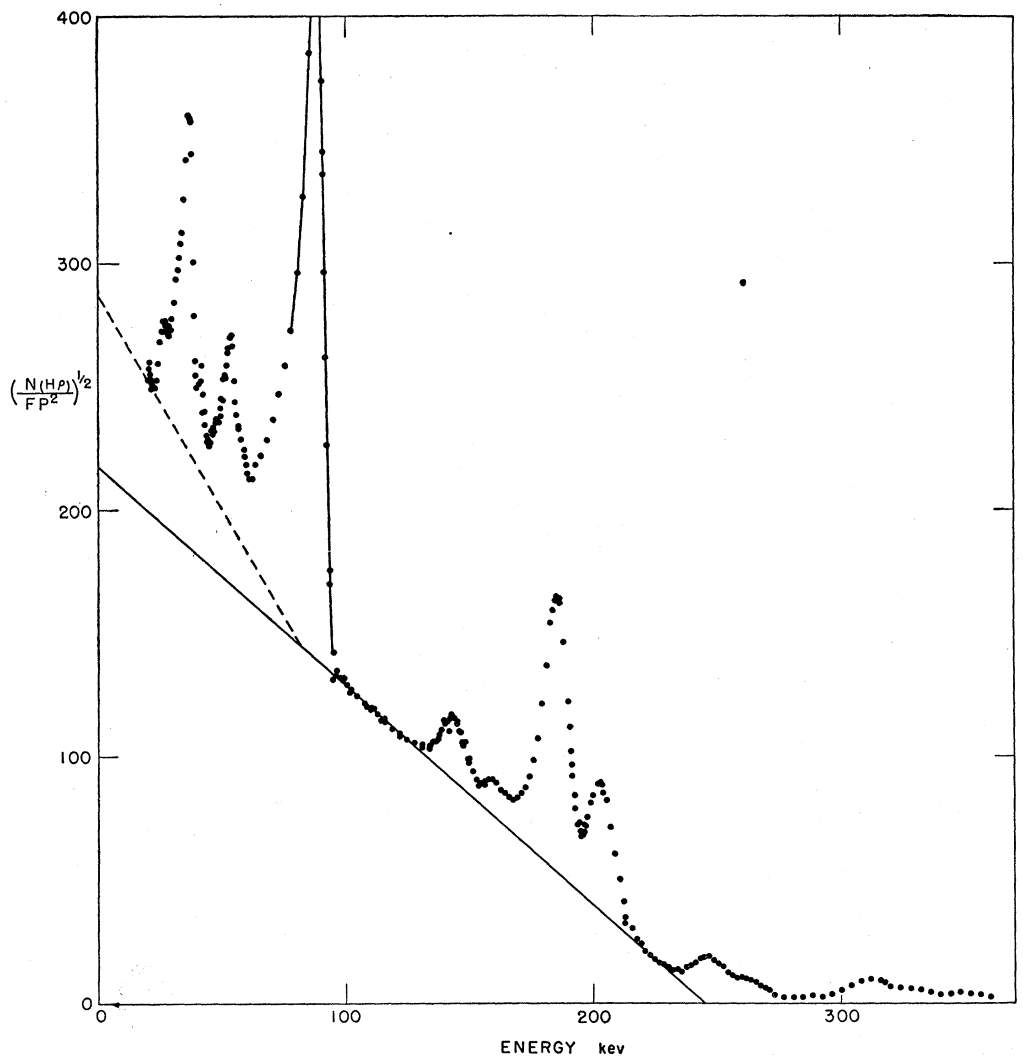

FIG. 1. Kurie plot of electron spectrum of U^{237}.

kev a thin window (*ca* 60 μg/cm^2) supported by a 50 percent transmission grid was used; above 95 kev a 1.3 mg/cm^2 mica window was used without the supporting grid.

The window transmission characteristics were determined with the use of a sample of Pm^{147} of average thickness <5 μg/cm^2. This sample was vacuum volatilized onto a 10-μg/cm^2 LC-600 film as a chelation compound of prometheum with trifluoro-acetylacetone [$Pm(F_3C_5H_4O_2)_3$]. It has been shown[8] that the Kurie plot of such thin samples of Pm^{147} is straight to less than 8 kev. The Kurie plots of our samples then indicated that the 60-μg/cm^2 window did not affect the shape above 20 kev, and the 1.3 mg/cm^2 above 80 kev. The two sets of data were normalized over the range (95 to 135 kev) where the slope of the momentum plot is low. The normalizing factor was determined at ten separate points in the overlap region. No single value of this factor varied by more than 1.5 percent from the average value.

The data from the beta-ray spectrometer show the presence of at least one beta-continuum with superimposed conversion lines. The Kurie plot (Fig. 1) of the beta-continuum, although masked for a considerable section owing to intense conversion lines, appears to be straight from an end point of 245±5 kev back to approximately 95 kev. Beta-gamma coincidence measurements on a scintillation sepectrometer (see below), which avoid the obscuring effects of the conversion lines, show a beta-component of 255±10 kev, of allowed shape. In this coincidence spectrum, the upturn in the Kurie plot (Fig. 2) observed below 100 kev corresponds to that normally seen in scintillation spectrometry of beta-rays, and so cannot be taken as evidence for the presence of a low energy beta-component.

Below *ca* 95 kev the continuum found in the lens spectrometer is masked by a number of conversion electrons as well as by distortion from source thickness

[8] Langer, Mott, and Price, Phys. Rev. **77**, 798 (1950).

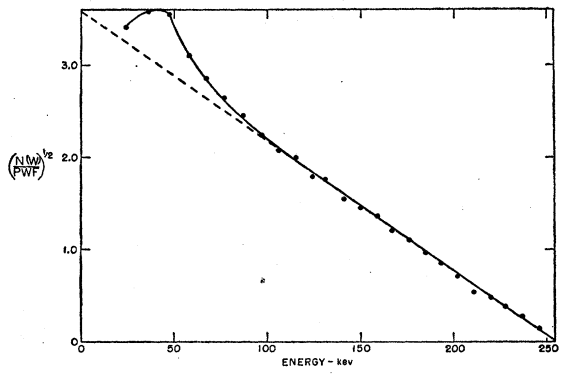

FIG. 2. Kurie plot of electron spectrum of U^{237} in coincidence with 208-kev gamma.

effects. The magnitude and sharpness of the rise below this energy (Fig. 1), in consideration of the source thickness, suggests strongly the presence of a beta-component of energy, say 80 to 95 kev (consistent with Slätis's observations). The only self-consistent decay scheme compatible with the gamma-energies found (see below) requires the existence of a beta of 81-kev maximum energy. If the sharp upturn in the Kurie plot below 95 kev is treated as adequate evidence for an 81-kev beta, analysis of the Kurie curve in the usual manner leads to beta branching ratios of $\gtrsim 80$ percent for the 245-kev, and $\lesssim 20$ percent for an assumed 81-kev beta (see dotted line, Fig. 1). However, it may be noted (see discussion) that calculations based on gamma-intensities furnish a minimum value for the branching ratio (*ca* 5 percent) for the 81-kev beta.

A higher energy beta-component was searched for without avail using a source of approximately three times the activity of the source used for the main study. The data indicated that the upper limit for the intensity of a beta of 511-kev energy (ground-to-ground transition for the suggested decay scheme, Fig. 6) was 0.1 percent of the intensity of the 245-kev beta.

The conversion lines (Table I) are assigned to levels in Np^{237} corresponding to gammas of 59, 165, 207, 334, 43, 269, 370, and 430 kev. The first four have been found in the gamma scintillation spectrometer. Careful search failed to reveal any line in the neighborhood of 118 kev, or of K_α Auger lines, contrary to the results of Slätis.[5] Values for the energies of the shell edges were obtained from unpublished tables of Hill *et al.*[9]

The K conversion line of the 334-kev gamma at 215 kev would not be observable due to the high intensity of the M conversion line of the 207-kev gamma at 201 kev. Similarly the K conversion line of the 269-kev gamma is masked by the L conversion line of the 165-kev gamma.

The K conversion line of the 165-kev gamma falls on the tail of line $M_I(59)$; in each experiment a discernible fluctuation was observed at 47.3 kev (750 $H\rho$), which is therefore assigned as the $K(165)$ line. Its maximum intensity, estimated by unfolding the tail of line $M_I(59)$, using as a guide the tailing edge of line $K(207)$ is 0.2 of the intensity of $L_I(165)$, which indicates that the multipolarity of the transition is at least 2^3.[10]

The K line of the 370-kev gamma is not resolved from the L line of the 269-kev gamma, but it is evidenced by a slight but reproducible decrease in slope at point $K(370)$ in Fig. 3.

A momentum plot of the spectrum is given in Fig. 3. The data from the lens spectrometer studies are summarized in Table I.

GAMMA-RAY STUDIES

To supplement the lens spectrometer data and aid in the formulation of a decay scheme a scintillation spectrometer and an argon-CO_2 proportional spectrometer were used to study the gamma ray spectrum.

A sodium iodide (thalliated) crystal in the scintillation spectrometer which had been calibrated with the 282-kev line of Hg^{203} [11] (resolution 17 percent), was

TABLE I. Summary of data of electron conversion lines and of gammas.

Lens spectrometer data				Scintillation spectrometer data		Transition data	
Conversion electron energy; kev	Shell converted in	γ energy kev	Relative[a] intensity	γ energy kev	Relative[b] intensity	Best value of energy; kev	Relative[c] intensity
...	...	...	...	27	...	27	
20.5	L_I	42.9	...	...	...	43	
26.7	L_{III}	44.3	...				
36.9	M_I	42.6	...				
41.7	N_I	43.2	0.013				
36.9	L_I	59.3	0.15	60	0.37	59	0.58
53.5	M_I	59.2	0.05				
57.6	N_I	59.1	...				
...	...	...	...	102[d]	0.53	102	0.53
47.3	K	165.5	...	163	...	165	
143.1	L_I	165.5	0.025				
159.1	M_I	164.8	0.003				
88.5	K	206.7	0.53	210	0.21	207	0.88
185.1	L_I	207.5	0.11				
202.5	M_I	208.2	0.03				
246.7	L_I	269.1	0.0022	...	...	269	
263.6	M_I	269.3	0.0007				
312.0	L_I	334.4	0.0007	340	0.025	334	
327.9	M_I	336.6	0.0002				
~251	K	~369.2		...	...	370	
348.0	L_I	370.4	0.00011				
312	K	430.2	...	...	...	430	

[a] Relative intensity = conversion electrons per 245-kev β.
[b] Relative intensity = gammas per 245-kev β.
[c] Relative intensity = conversion electrons plus gammas per 245-kev β.
[d] K x-rays. (Assumed.)

[9] Hill, Church, and Mihelich, Rev. Sci. Instr. **23**, 523 (1952). We wish to thank the authors for a pre-publication copy of these tables.

[10] M. Goldhaber and A. W. Sunyar, Phys. Rev. **83**, 906 (1951).
[11] H. Slätis and K. Siegbahn, Phys. Rev. **75**, 318 (1949).

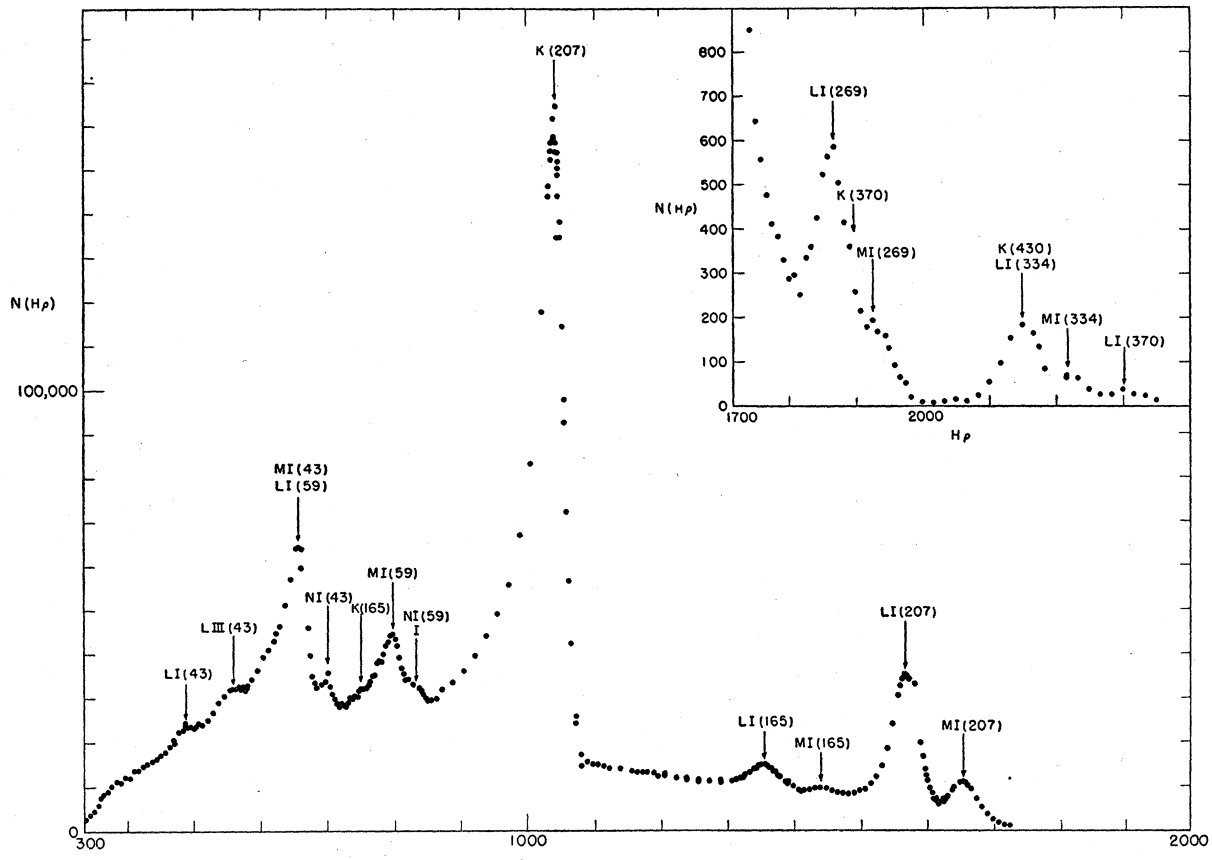

FIG. 3. Momentum plot of electron spectrum of U^{237}. (Note change of scale on inset.)

used. Gammas of approximately 60, 210, and 340 kev were found (Fig. 4). A search for gammas of higher energy than 340 kev was carried out using very intense source with lead absorbers (*ca* 1.9 g/cm^2) to keep the *ca* 210-kev gamma from flooding the counter. No gammas were found at these higher energies although the presence of these gammas in intensity 10 percent that of the *ca* 340-kev gamma would have been observable.

The argon-CO_2 counter was used to examine the quantum spectrum below 60 kev. The K x-rays of silver (22.1 kev) were used to calibrate this instrument which gave a resolution width of 14 percent at this energy. The results showed a large amount of L x-ray present in the U^{237} spectrum but no evidence for the existence of the 43-kev gamma found from the conversion line spectrum. However, Prohaska[12] has reported the presence of an approximately 100 percent converted 42-kev gamma in the α-decay of Am^{241} (see discussion). The 27-kev gamma did not show up directly but could have been lost in the tailing out of the x-ray lines whose counting rates were significantly above background at this setting. The results of these studies (including estimates of relative gamma-ray intensities) are listed in Table I.

COINCIDENCE STUDIES

Two sodium iodide (thalliated) scintillation spectrometers were used in coincidence to study the energy level structure of the Np^{237} product nucleus.

With one set to count the 210-kev gamma, a survey of the energy spectrum with the other showed coincidence with gammas of energies 27, 60, and 165 kev (Fig. 5). The 165-kev gamma was hidden under the intense 210-kev line in the singles measurement.

A search for gammas in coincidence with the 340-kev gave negative results.

Substituting an anthracene crystal beta-counter for one of the gamma counters showed the 210-kev gamma to be in coincidence with a beta of an end-point energy of 255±10 kev (Fig. 2).

An attempt to find coincidence between the 340-kev gamma- and beta-rays was not successful owing to the limitations on the total singles counting rates of the instruments available.

[12] C. A. Prohaska, University of California Radiation Laboratory Report UCRL-1395 (1951) (unpublished); Asaro, Reynolds, and Perlman, Phys. Rev. **87**, 277 (1952); F. Asaro *et al.* from their α-spectrum data in the decay of Am^{241} construct a decay scheme which leads to a ground state in Np^{237} 11 kev lower than that reported here. Consequently in referring to energy levels in Np^{237}, their designation of any particular level differs from ours by 11 kev. The 11-kev transition has not been observed.

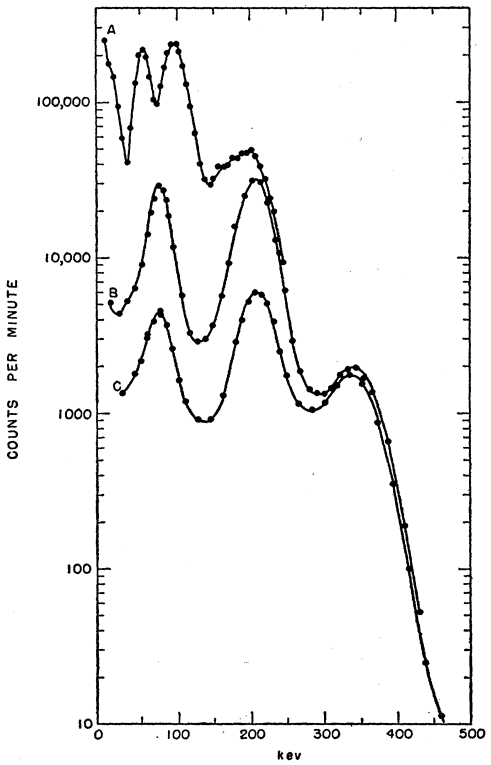

FIG. 4. Gamma-spectra of U^{237} in sodium iodide scintillation spectrometer. [The vertical position of the curves with respect to each other has no significance. The peak on curves *B* and *C* at *ca* 75 kev is lead x-ray.] A. 0.00-g/cm² lead absorber; B. 1.10-g/cm² lead absorber; C. 4.00-g/cm² lead absorber.

SUMMARY AND CONCLUSIONS

A decay scheme (Fig. 6) has been assembled from the data of the several experiments performed on this isotope. Many of the features of this scheme are only tentative; however, it is felt that the main features are well established and that the total decay energy of U^{237} is correctly defined. This energy is given by the 245-kev beta in cascade with the 207- and 59-kev gammas; this is the only plausible scheme in view of the intensity and coincidence measurements.

The position of the 165-kev gamma is fixed by the fact of its coincidence with the 207-kev gamma, and by its low intensity which indicates that the intense 268-kev level[12] cannot feed it.

The fitting together of other features of the decay scheme is somewhat tenuous.

Studies of the fine structure[12] in the alpha-decay of Am^{241} have given some information about the energy level structure of Np^{237}. The results of these studies indicate a level at 59 kev[12] and one at approximately 100 kev,[12] the 59- and 100-kev gammas having been observed. This data has been incorporated into our decay scheme.

The scintillation spectrometer showed a line at 102 kev which could well be a mixture of *K* x-rays and *ca* 100-kev gammas. The *L* conversion line from a 100-kev gamma would fall on the side of the *K* line from the 210-kev gamma and was not seen (see text). The results of the coincidence experiments indicate that the 334-kev gamma is not all in cascade with the 59-kev gamma, since a crude calculation shows that, had this been the case, the coincidences would have been numerous enough to have been observed. This indicates that the 100-kev gamma must be comparable in intensity to the 334-kev gamma (*ca* 3 percent of the 245-kev β).

The intensity values given in Table I are based on the assumption that all of the 102-kev quanta found in the scintillation spectrometer are *K* x-rays arising from the conversion of the 207-kev gamma, and that there is one *K* x-ray per *K* electron; that is, small corrections for *K* fluorescent yield are neglected.[13]

These assumptions are based on the following facts: (1) The energy measured is in reasonable agreement with predicted K-α x-ray energies (97 to 101 kev); (2) From the beta-spectrum data it is known that the 207 gamma is *ca* 50 percent *K* converted; (3) No gamma of 102 kev was found in coincidence with the 207-kev gamma, thus eliminating such a gamma from the main cascade of the U^{237} nucleus and consequently eliminating it as a component of any important intensity (see Fig. 6). Although the relative intensity of the 59-kev transi-

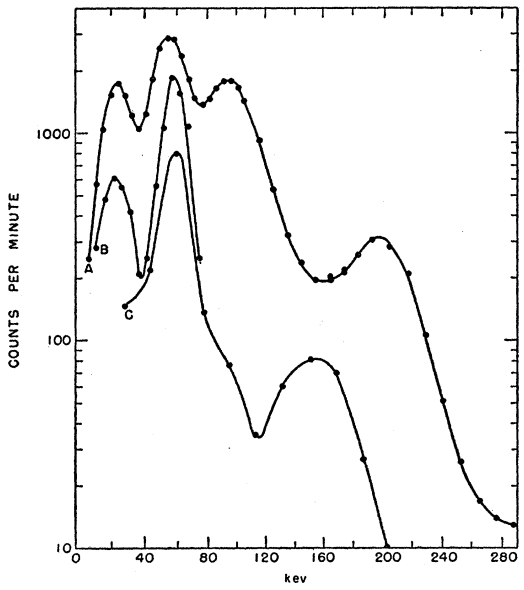

FIG. 5. Gamma-gamma coincidence spectra of U^{237} in sodium iodide scintillation spectrometers. [The vertical position of the curves with each other has no significance.] Abscissas denote energy setting of spectrometer I; coincidences observed with: A. Gammas above 20 kev in spectrometer II; B. 200-kev gammas in spectrometer II, 1.0 g/cm² of copper absorber over counter II; C. 200-kev gammas in spectrometer II, 1.4 g/cm² of copper absorber over each counter.

[13] Extrapolation of the data given by A. H. Compton and S. K. Allison [*X-Rays in Theory and Experiment* (D. Van Nostrand Company, Inc., New York), second edition, p. 488] leads to estimates for the *K*-fluorescence yields in heavy isotopes of within a few percent of unity.

tion (Table I) is lower than that of the 207-kev, it is evident that it must be in sequence with the 207-kev transitions, and from the data on the Am^{241} alpha-decay[12,14] it is located below the latter.

The energy level scheme proposed by Asaro *et al.*[12] based on alpha- and gamma-decay studies on Am^{241} includes 26.43- and 33.36-kev transitions in sequence, in parallel with the 59-kev transition (shown dotted in Fig. 6), and with intensities of the unconverted gamma one-third that of the 59-kev gamma. As noted above the 27-kev was found in coincidence with the 207-kev gamma, although the 33-kev gamma was not seen. We have also observed the 27-kev gamma in Am^{241} [14] but not the 33-kev gamma, on a scintillation and on a proportional counter spectrometer, in agreement with the results of Beling *et al.*[15] (Fig. 7). We did not survey the electron spectrum to low enough energies (<20 kev) to find the L lines of 26.4- or 33.3-kev gammas. On the assumption that the results of Asaro *et al.*[12] on the existence and intensity of the 33-kev gamma are correct,

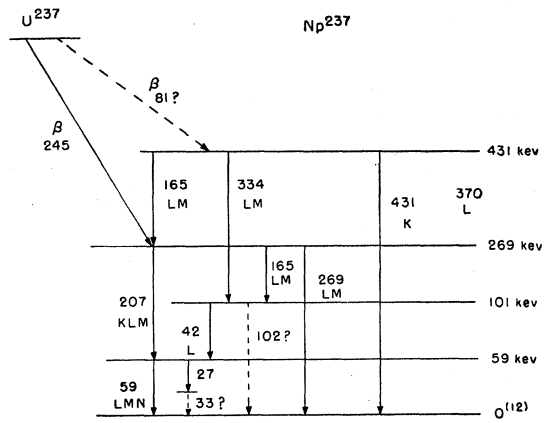

FIG. 6. Decay scheme of U^{237}.

quite reasonable values of the conversion coefficients of the 27- and 33-kev transitions lead to a matching of the intensities of the 207-kev transitions with the intensities of the transitions (59, 27, and 33 kev), which deplete the 59-kev level), and thus affirm the validity of the proposed decay scheme.

The K conversion coefficient of the 207-kev transition and the K/L conversion ratio have been calculated from the lens spectrometer data. An attempt to correlate these values with the character and multipolarity of the 207-kev gamma predicted by the K/L conversion ratio and by the K conversion coefficient was unsuccessful. According to the tables of Rose *et al.*[16] the K conversion coefficient ($\alpha=1.6$) corresponds to an $E4$ transition ($E4=1.1$, $E5=2.3$, $M1=4$, $M2=10$) whereas according

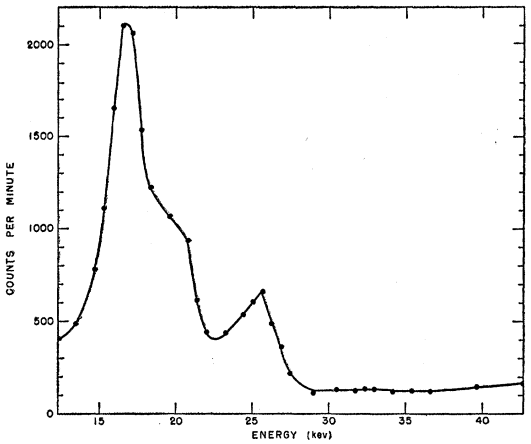

FIG. 7. Gamma-spectrum of Am^{241} taken with argon$-CO_2$ proportional counter. 440 mg/cm² Al over sample.

to the work of Goldhaber and Sunyar[10] the K/L conversion ratio (5) corresponds to an $M2$ transition (Goldhaber and Sunyar values: $M1=7.8$, $M2=5$, $M3=2$, $E1=3$, $E2-E4<1$).

The branching ratio between the 245-kev beta and 81-kev beta can be estimated from the Kurie plot and from the intensities of the gammas in the decay of the 431-kev level of the Np^{237} nucleus (Fig. 6). The maximum amount of 81-kev beta possible on the basis of the Kurie plot is approximately 20 percent of the total betas. The minimum amount necessary to account for the intensity observed for the 165-kev gamma (+conversion lines) is of the order of 5 percent.

On the basis of a 20 percent branching to an 81-kev beta and 80 percent to a 245-kev beta, the logft values for these two betas are 5.9 and 6.6, respectively; for a 5 percent and 95 percent division the logft values become 6.5 and 6.5.

A value of 511 kev for the decay energy of the U^{237} leaves a residual unbalance of *ca* 160 kev in the following decay cycle if one assumes the alpha-decay energy for U^{237} to be 4.28 (larger than the 4.25-Mev alpha-decay energy of U^{238}).

$$\begin{array}{ccc} & \overset{\alpha}{\longleftarrow} & \\ Pa^{233} & & Np^{237} \\ \beta^- \uparrow & \alpha & \uparrow \beta^- \\ Th^{233} & \longleftarrow & U^{237} \end{array}$$

However, work is being carried out at this laboratory[17] searching for gammas in cascade with the Np^{237} alphas, and preliminary experimental results indicate that there is gamma-energy of the required order of magnitude to account for the unbalance in sequence with the alpha-decay.

Therefore, although many features of the presented decay scheme are uncertain, these experiments serve to determine the decay energy of U^{237}.

[14] Freedman, Wagner, and Engelkemeir, Phys. Rev. 88, 1155 (1952).
[15] Beling, Newton, and Rose, Phys. Rev. 86, 797 (1952).
[16] Rose, Goertzel, and Perry, Oak Ridge National Laboratory Report ORNL-1023 (1951) (unpublished).
[17] L. B. Magnusson and D. W. Engelkemeir (private communication).