The Radiations of Th^{231}(UY)

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β-ray spectrum studies on a double-lens spectrometer of the electron spectrum of UY revealed a complex spectrum with three beta-components of 302 kev (44 percent), 216 kev (11 percent), and 94 kev (45 percent), and numerous conversion lines assignable to gammas of 22, 59, 63, 85, 122, 167, and 208 kev, each of these except the 122 kev also observed directly as gammas in a scintillation spectrometer. There is also evidence for a 107- and for a 230-kev gamma-ray. The samples were separated carrier-free from isotopically pure U^{235} (99.86 percent) by solvent extraction. A decay scheme is presented which gives a total disintegration energy of 324 kev.

INTRODUCTION

ONE of the naturally-known beta-emitters is Th^{231} (or UY), being the daughter by alpha-decay of U^{235}. It has also been formed by the reactions $Th^{230}(n,\gamma)Th^{231}$ [1] and $Th^{232}(n,2n)Th^{231}$.[2]

Most of the previous measurements of the radiations of UY which have been reported were performed on samples milked from the U^{235} present in natural uranium. Due to the decay $U^{238}\xrightarrow{\alpha}UX_1(Th^{234})$, the milked UY samples contained large amounts of both UX_1 and its daughter $UX_2(Pa^{234})$. The presence of these interfering activities greatly complicated experiments on UY radiations, since the isotopic abundance of U^{235} is only 0.7 percent. The availability of enriched U^{235} with attendant reduction of the UX_1 content, made possible more clear-cut measurements. UY from such sources has been investigated by absorption techniques,[3,4] with the relatively poor resolution of such methods. The results indicated that the radiations of UY included: (1) a single beta-ray of about 0.2 Mev; (2) *L* x-rays, presumably from conversion of low energy gamma-rays; (3) a very soft component presumably due to *M* x-rays and low energy conversion electrons; (4) several gamma-rays, whose energies were 35 kev, 65 to 75 kev, and 100 kev. The last two gamma-rays were uncertain as to energy.

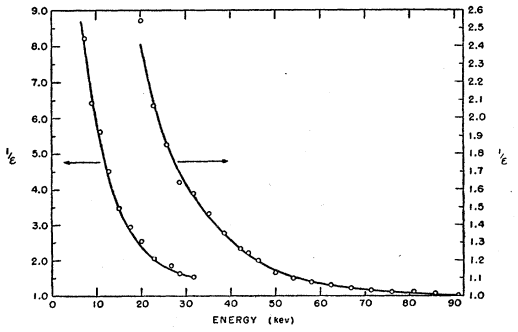

FIG. 1. Efficiency (ϵ) of anthracene scintillation counter as a function of energy.

More recently, the radiations from UY extracted from natural uranium have been examined with a beta-ray spectrometer.[5] The results indicated the presence of gamma-rays at 59, 63, 82, and 120 kev; the beta-ray end point was found to be 390 kev, with some indication of lower energy branches with 190- and 100-kev maximum energies.

In the work reported here,[6] an enriched U^{235} sample was used to supply the UY source. The radiations were examined with a magnetic beta-ray spectrometer and a gamma-ray scintillation spectrometer. Sufficient information was secured to make possible the proposal of a decay scheme which fits the measurements reasonably well.

EXPERIMENTAL—ELECTRON SPECTROMETRY

A double lens spectrometer adjusted for a transmission of *ca* 2 percent with a resolution of *ca* 3 percent was used to examine the electron spectrum of UY. The spectrometer calibration was based on a value of 624 kev[7] for the *K* conversion line of Ba^{137}.

An anthracene scintillation counter served as a detector, the scintillations being detected by a 1P21 photomultiplier at liquid nitrogen temperature, located in a magnetic shield outside the magnetic field of the spectrometer lens coils. The light flashes from the 1 mm thick by 8 mm diameter anthracene crystal were piped to the photomultiplier through a 20 inch long$\times\frac{3}{4}$ inch diameter clear quartz light guide, to which the crystal was cemented.

The efficiency of this system for detection of electrons fell below 100 percent for energies less than 80 kev. A determination of the efficiency correction applicable to the counting data was made by a comparison of the Kurie plot of an experimental spectrum of Pm^{147} to the known allowed shape. A very thin (<10 $\mu g/cm^2$) sample of Pm^{147}, prepared by vacuum volatilization of the

[1] A. H. Jaffey and E. K. Hyde, Argonne National Laboratory Report ANL-4249 (1949) (unpublished).
[2] Nishina, Yasaki, Kimura, and Ikawa, Nature **142**, 874 (1938).
[3] Jaffey, Lerner, and Warshaw, Phys. Rev. **82**, 498 (1951). Includes survey of previous work.
[4] G. B. Knight and R. L. Macklin, Phys. Rev. **75**, 34 (1949).
[5] Stoker, Hok, and Sizoo, Physica **17**, 164 (1951).
[6] Freedman, Jaffey, May, and Wagner, Argonne National Laboratory Report ANL-4613, 67 (1951) (unpublished); Phys. Rev. **86**, 633 (1952).
[7] L. Langer and R. D. Moffat, Phys. Rev. **78**, 74 (1950).

trifluoro-acetylacetonate compound onto a *ca* 10 $\mu g/cm^2$ LC-600 film, gave a Kurie plot linear to *ca* 80 kev, but dropped below the linear extrapolation at lower energies, down to <8 kev. Such thin samples have been shown[8] to give an allowed shape down to *ca* 8 kev. A graph of the efficiency correction factor is given in Fig. 1.

Counting data was recorded automatically on a chart recorder by a logarithmic counting rate meter, as the spectrometer coil current was swept continuously over the necessary range. The counting rate was determinable to ±1 percent from the chart trace, and the current at any chart position to within ±0.1 percent.

The spectrographic data reported here are the results of several experiments made on separate samples, each the product of an individual chemical separation. The UY samples were prepared by chemically isolating the UY (in radioactive equilibrium) from a stock solution of 25 grams of U^{235} [9] in HNO_3. The chemical separation, carried out for us by Paul Fields, Gray Pyle, and Ruth Sjoblom of this Laboratory, was designed to yield samples without carrier and with a minimum of massive impurities present. The thorium was separated from the uranium, as well as from impurities, by making use of the formation of a complex with thenoyl-trifluoro-

TABLE I. UY—Beta-components.

Maximum energy of beta (kev)	Relative intensity (%)
302±2	44
216±5	11
94±2	45

acetone (TTA) in benzene solution. The thorium was further purified by usual well-known chemical procedures, with particular attention to the purity of the reagents involved, and to the thorough destruction of residual organic matter. The purified thorium was obtained as a minute dry deposit of the chloride in a platinum crucible.

The almost invisible deposit was taken up in 5λ of pure 3*N* HCl solution and deposited on a thin (<20 $\mu g/cm^2$) film of LC-600 on a circle of *ca* 5 mm diameter. The amount of impurity coming through the chemical procedure varied among the several experiments performed; the spectrometer results clearly showed the presence of impurities which increased the source self-absorption in terms of a broadening of the conversion line peaks, becoming worse with decreasing energy. The best experiments were done with samples estimated to be of average thickness 20 $\mu g/cm^2$, fairly uniformly spread over the sample area. Similar sources were used in the scintillation gamma-spectrometer experiments described below.

[8] Langer, Motz, and Price, Phys. Rev. **77**, 798 (1950).
[9] Obtained from Union Carbon and Carbide Corporation, Oak Ridge National Laboratory. Isotopic analysis—99.86 percent U^{235}; 0.07 percent U^{238}; 0.07 percent U^{234}.

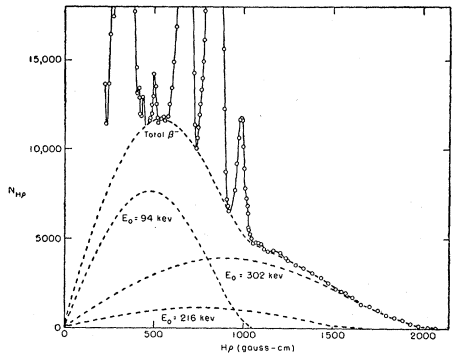

FIG. 2. Momentum spectrum of Th^{231} (UY).

From the amassed data the three runs which clearly gave the best results were taken as a basis for computation. Figure 2 presents an over-all momentum spectrum and Fig. 3 a Kurie analysis. Corrections for counting efficiency and for decay of the sample have been applied.

Successive subtractions of apparently linear components from the Kurie curve indicated beta-continua (Table I) of energies $E_0=302\pm2$ kev (44 percent relative abundance), $E_0=216\pm5$ (11 percent) and 94±2 kev (45 percent). No beta of intensity >2 percent of the 302-kev beta and of substantially higher energy was observed, in contradiction to the results of Stoker *et al.*[5] The relative abundance of the low beta-component is subject to considerable error, inasmuch as imperfectly resolved conversion lines overlie the major portion of the spectrum. A short section of the curve between 23 and 28 kev [between lines $N_{IV}(1)$ and $L_I(2)$] seems well resolved from the tail of line $L_I(2)$ and to have no other lines on it, and so was considered as reliable to guide the drawing in of the low beta; similarly for the regions at 45 kev and 70 kev. A further indication of the correctness of this subtraction is had from Fig. 4, where the profiles of the conversion lines are shown on a momentum plot after subtracting the 94-kev beta. The line shapes are similar to those normally obtained with this instrument, especially in their low energy tails; subtraction of a beta of much

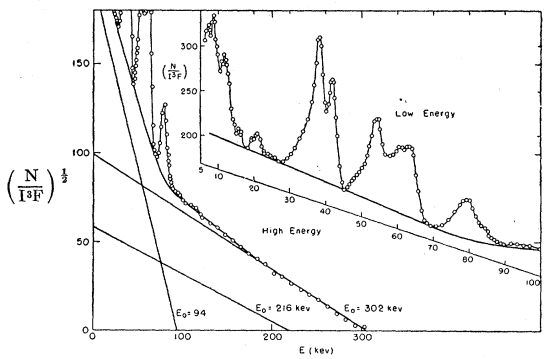

FIG. 3. Kurie plot for Th^{231} (UY).

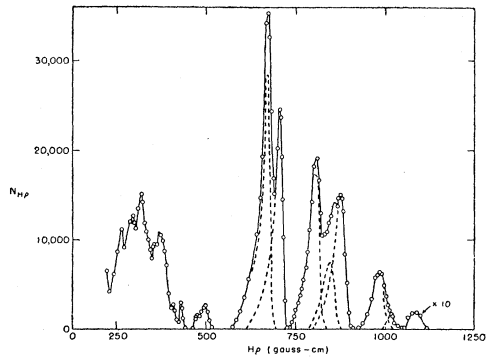

FIG. 4. Momentum plot of conversion lines of Th^{231} (UY); beta-continue subtracted off.

lower intensity would result in excessively large base widths of the lines, compared to their half-widths [3.6 percent for line $L_I(2)$]. However, as will be seen in the discussion below, a major difficulty with our interpretation of the experimental results is associated with an apparently too high branching ratio for the low energy beta.

Table II summarizes the information on the conversion lines. Many of the lines are low in intensity, and lie in an energy region where the counter efficiency is low. To distinguish statistical fluctuations from true lines, each of three selected data charts was examined. Only lines which appeared on all three were considered.

The unfolded shapes of incompletely resolved lines were obtained by construction, using leading and trailing edge profiles of nearby resolved lines as a guide. From these shapes the relative line intensities and peak locations were obtained graphically. No attempt was made to resolve the group below 14 kev completely, except to obtain a rough value for the relative intensity of line $K(5)$, which is an upper limit estimate. No lines were observed at 98 or 115 kev, contrary to the observations of Stoker *et al.*[5]

In columns I, IV, and V of Table II are given the line assignments. The shell binding energies were obtained by a Moseley law interpolation from data[10] for neighboring elements. Of the 18 lines definitely observed, the assignment of eight seems certain (labeled *) and of nine, probable (labeled ↑). One line (**) was not assigned. In some cases the same line is assigned to two levels, using the intensities and provisional assignments of other lines and the results of the gamma-ray spectroscopy (see below) as a guide. Where a line is so assigned, the major component of course determines the observed line energy, and a small unresolved component may be assigned with some latitude on the energy scale; the actual peak values were used in the assignments and perhaps account for some of the disagreements in energy between various conversion lines assigned to one level.

[10] A. H. Compton and S. R. Allison, *X-Rays in Theory and Experiment* (D. Van Nostrand and Company, Inc., New York, 1935), second ed., p. 794.

Line $K(6)$ was assigned to a 208-kev transition on the basis of the results of gamma spectroscopy. Some of the lines assigned to the 22-kev gamma could equally well have been allotted to gammas of 26 or 28 kev; 22 kev was chosen as giving the best energy balance in the proposed decay scheme, and in agreement with the results of gamma-ray spectroscopy (see gamma-ray measurements, below). In fact, the relative intensities of lines $M_I(1)$, $M_{III}(1)$, and $N_{IV}(1)$ suggest that the assignment may not be correct, and that 25–26 kev would be more reasonable. Lines A_1, A_2, A_3, and A_4 agree satisfactorily with predicted Auger transition energies, although considering the poor resolution and the large number of possible Auger transitions in this energy region such allocations are not very significant. Line $K(5)$ was ascribed to a cross-over transition of 122 kev spanning the 59-kev and 63-kev transitions and $K(7)$ is ascribable, as a small fraction of line $M_{II}(2)$, to a cross-over of the 63-, 85-, and 22-kev gammas.

In column VII of Table II are listed the intensities of the conversion lines relative to the intensity of the beta plus gamma and internal conversion transitions assumed to feed the emitting level in question, according to the decay scheme proposed in Fig. 7 (see discussion below). The conversion line intensities relative to the sum of all beta transitions appear in column VI.

TABLE II. UY conversion lines and gammas.

I	II	III	IV	V	VI	VII
Line	Energy (kev)	Binding (kev)	Gamma (kev)	Gamma best value (kev)	Line or gamma-intensity relative to total beta intensity	Line or gamma-intensity relative to intensity of betas feeding level
$L_{III}(1)$ ↑	6.0	16.8	22.8			
$M_I(1)$ ↑	16.0	5.4	21.4			
$M_{III}(1)$ ↑	17.6	4.2	21.8			
				22	0.80 − [0.5(γ) +0.3(*L* x-rays)]	
$L_I(2)$*	38.2	21.2	59.4		0.094	0.209
$M_{II}(2)$*	54.3	5.1	59.4		<0.066	<0.147
$N(2)$*	59.6	1.	60.6			
				59.4	0.063	0.15
$L_I(3)$*	41.9	21.2	63.1		0.083	0.184
$M_{IV}(3)$*	59.6	3.6	63.2		0.031	0.069
$N(3)$*	63.7	1.	64.7			
				63.1	0.126	0.28
$L_I(4)$*	63.7	21.2	84.9		0.060	0.107
$M_I(4)$*	79.4	5.4	84.8		0.026	0.046
$N_{IV}(4)$*	84.0	0.7	84.7		0.004	0.008
				84.8	0.44 (normalized)	0.79
				107		
$K(5)$ ↑	8.7	113.1	121.8		<0.06	<0.13
				121.8	0.008	0.018
$K(6)$*	94.9	113.1	208.0		0.0007	0.002
				208	0.0009	0.002
$K(7)$ ↑	54.3	113.1	167.4		≪0.066	≪0.147
				167.4	0.0066	0.014
				230	0.0004	0.001
**	10.9	(Unassigned)				
			Possible Auger transitions			
A_1 ↑	7.6	$L_{III}-M_I-M_{IV}=7.7$; $L_{III}-M_{II}-M_{III}=7.5$				
A_2 ↑	9.9	$L_{II}-M_I-M_{II}=10.0$; $L_{II}-2M_I=9.6$; $L_{III}-M_{III}-M_V=10.1$				
A_3 ↑	11.9	$L_I-M_I-M_{III}=11.6$; $L_I-M_I-M_{IV}=12.2$; $L_I-M_{II}-M_{III}=12.0$; $L_{II}-M_{II}-M_{IV}=11.8$; $L_{II}-2M_{III}=12.1$; $L_{III}-2M_V=11.8$				
A_4 ↑	14.7	$L_I-M_{III}-M_V=14.6$; $L_{II}-M_{IV}-M_V=14.3$.				

* Denotes line assignment considered certain.
↑ Denotes line assignment considered provisional.

From the conversion lines it was clear that there existed gamma-rays of 85, 63, 59, and 22 kev in considerable abundance. In addition, there was a very low intensity 95-kev conversion line $K(6)$, ascribed to K conversion of a 208-kev gamma-ray. These gamma-rays fitted into a consistent decay-scheme, since the differences in the beta-ray energies (maximum energies 93, 216, 302 kev) corresponded (Fig. 7) to the gamma-ray energies.[11] Most of the possible cross-over gamma-rays were not observed; the intensities of the conversion lines were either too low or the conversion lines resulting were hidden under the conversion lines of the more intense gamma-rays.

EXPERIMENTAL—GAMMA-SPECTROSCOPY

A search for unconverted gamma-rays was made, using the conventional photoelectric method with the beta-ray spectrometer. The results were negative because of low intensity resulting from weakness of sample activity and low conversion efficiency in the thin converters necessary at such low gamma-ray energies.

Since it was evident from preliminary measurements that a simple absorption counting technique could not handle the complex gamma-mixture involved, a NaI(Tl) crystal spectrometer[12,13] was used. A crystal $1\frac{1}{4}$ inches in diameter and $\frac{1}{2}$ inch thick was hermetically enclosed in an 0.030-inch thick aluminum cup (206 mg/cm^2) with a Pyrex glass disk sealed on the side facing the 5819 photomultiplier. An electrical tape (*ca* 50 mg/cm^2) light shield covered the entire crystal-photomultiplier combination. The sample was as close to the crystal as possible, at a geometry of about 30 percent. The output of the photomultiplier was linearly amplified and analyzed with a single channel differential pulse-height selector, with variable channel width. The channel-width was less than 10 percent of any peak width, and hence had a negligible effect upon the resolution.

Preliminary examination of a UY source (Fig. 5c) showed that while most of the activity lay in two peaks at 22 and 85 kev, there was evidence of the presence of gamma-rays at other energies, although these were not resolved. The shape of the low energy side of the 85-kev peak indicated that there might be a gamma-ray in this region, while the tailing off of the high energy side went far beyond the expected straggling of a single peak.

In order to decrease the relative concentration of the 85-kev peak, the peaks were differentially attenuated, by placing absorbers between sample and crystal. Appropriate absorbers were chosen in the following manner: A plot of half-thickness *vs* quantum energy for any particular absorber material characteristically has an increasing slope above the K-edge for some range of energy, followed by a decreasing slope. The maximum discrimination between two gamma-rays (for a given residual intensity) is achievable with the absorber

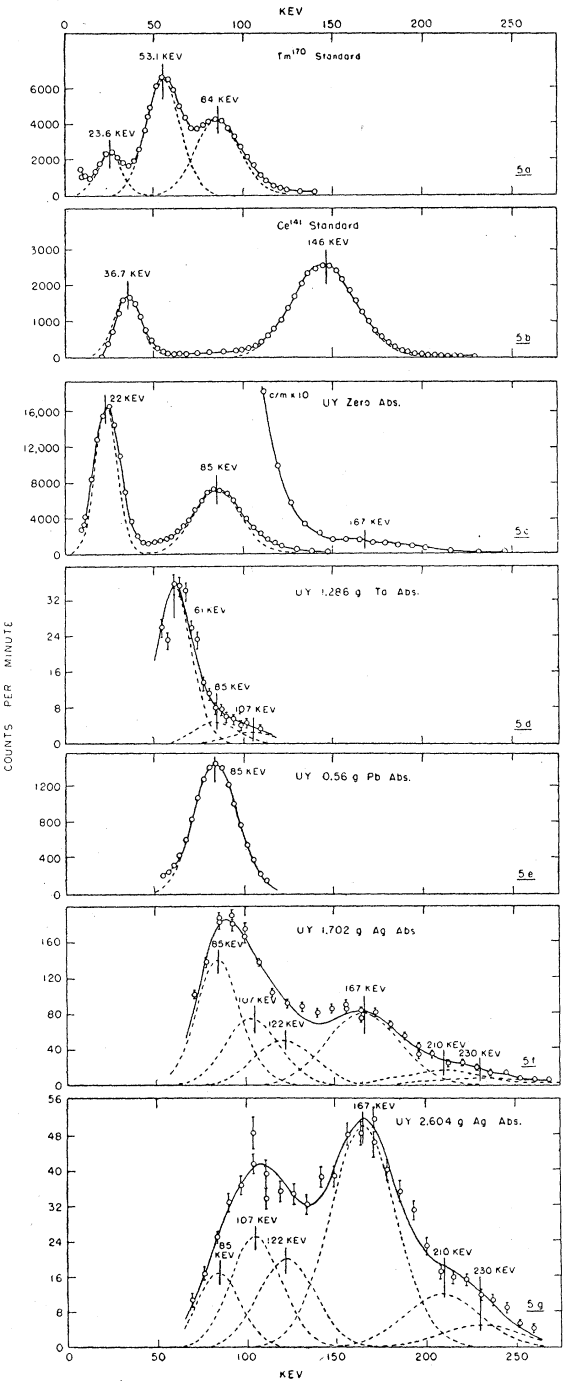

FIG. 5. Gamma-spectra of Th^{231} (UY) on scintillation spectrometer.

[11] 216−93=123; 63+59=122; 302−216=86; 302−93=209.

[12] This was an instrument developed by R. Swank of this laboratory [R. K. Swank, Argonne National Laboratory Report ANL-4303 (1949) (unpublished); R. K. Swank and J. S. Moenich, Rev. Sci. Instr. (to be published)], and was essentially similar to published versions (see reference 13) of NaI(Tl) gamma-ray spectrometers. We wish to thank Mr. Swank for the preparation of the NaI(Tl) crystal and for the design and construction of the amplifier and pulse-height analyzer circuits.

[13] S. A. E. Johannson, Nature **165**, 396 (1950); R. W. Pringle, Nature **166**, 11 (1950); Bell, Cassidy, and Kelley, Phys. Rev. **82**, 103 (1951); R. Hofstadter and J. McIntyre, Phys. Rev. **78**, 617 (1950); **80**, 631 (1950); Nucleonics **7**, 32 (September, 1950).

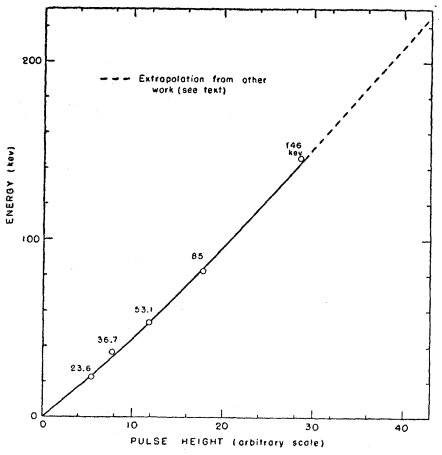

FIG. 6. Pulse amplitude *vs* energy for gammas on NaI(Tl) scintillation spectrometer.

material having the largest slope in the energy region of interest, and hence whose K-edge lies considerably below this region. Of the absorbers available, it was calculated that silver would give the best discrimination between an 85-kev gamma-ray and one with an energy somewhat above 100 kev, although a copper absorber would not be much worse. Using some estimates as to energies and relative intensities derived from the absorption measurements, it was calculated that a 1.7-g/cm^2 absorber would give the best discrimination. The pulse analysis is shown in Fig. 5f. It was evident that the 85-kev gamma-ray had not been discriminated against sufficiently, and a heavier (2.6-g/cm^2) silver absorber was used (Fig. 5g). Although the resolution was improved in this measurement, still greater discrimination would have been possible with a thicker absorber; unfortunately, the sample intensity was too low to allow still further attenuation.

Both curves clearly show the presence of two gamma-rays at about 100 kev and 165 kev, with some indication of at least one lower-intensity peak above 200 kev. The detailed analysis of the two curves is discussed below.

In order to examine the region covering the low energy wing of the 85-kev peak, preferential absorption was again used. This involved absorption by an element whose K-edge lay below 85 kev, since the smallest half-thickness (or greatest absorption) occurs just above the K-edge. The shape of the curve in Fig. 5c indicates the possible presence of the 59- and 63-kev gamma-rays indicated in the β-spectrometer curves; thus the best discrimination would not occur with the elements whose K-edge lies just below 85 kev [mercury, $K=83.2$ kev and gold, $K=80.9$ kev[10]] since the half-thickness decreases very rapidly below the K-edge. Of the easily available elements, tantalum was chosen as the best material, and Fig. 5d shows a curve taken with a 1.3-g/cm^2 tantalum absorber.[14] Very good discrimination was achieved, with a 61-kev gamma-ray dominant and only a small tail resulting from the 85-kev (and higher) gammas. The resolution was too poor to separate 59- and 63-kev gamma-rays, if both were present.

A lead absorber (K-edge$=88.2$ kev) did not attenuate the 85-kev gamma-ray very much, but did attenuate the gamma-rays above and below it in energy, so that Fig. 5e shows a practically "clean" peak at 85 kev.

Standardization of pulse height in terms of energy was achieved by the use[15] of Ce^{141} and Tm^{170} as standards (Figs. 5a and 5b).

The energy of the Ce^{141} gamma-ray is 146 kev,[16] while that of Tm^{170} is 85 kev.[17] In addition to the gamma-rays, the K x-rays of the daughter elements, praseodymium (Ce^{141} decay) and ytterbium (Tm^{170} decay) were observed, since both gamma-rays are partially converted. Since the scintillation spectrometer does not resolve the K_α and K_β lines, averaged[18,19] values for the effective K x-ray energies were taken, 36.7 kev for Pr and 53.1 kev for Yb.

For softer quantum radiation, the stopping power of NaI is so great that the absorption occurs close to the surface of the crystal; some of the iodine K x-rays following the photoelectric process escape backwards through the crystal surface, resulting in the formation of the "escape peak."[20] This peak occurs at an energy corresponding to the difference between the quantum radiation energy and that of the iodine K x-rays (suitably averaged). The effect is small at higher energies—only 6 percent at 85 kev,[21] so that the only escape peak observable was that due to the Yb K x-rays, at 23.6 kev. The escape peak (55.8 kev) from the Tm^{170} 85-kev gamma-ray occurred at the high energy side of the Yb K x-ray peak and hence was unobservable. The two gamma-ray lines, the two K x-rays and the escape peak were used to construct a pulse-height-energy calibration curve (Fig. 6).

Examination of the high energy tail of the UY curves (Figs. 5f and 5g) showed some high energy components beyond the highest point (146 kev) on the calibration curve; their energies were evaluated using other calibration curves measured in this laboratory,[22] normalized to our curve. The calibration curve in Fig. 6 shows a distinct curvature indicating higher efficiency at lower photon energies. This characteristic has been observed

[14] Ta, K edge-67.5 kev.

[15] We wish to thank Donald Englekemeir and Theodore Novey for providing us with samples of these activities.

[16] H. B. Keller and J. M. Cork, Phys. Rev. **84**, 1079 (1951); M. Freedman and D. Engelkemeir, Phys. Rev. **79**, 897 (1950).

[17] J. S. Fraser, Phys. Rev. **76**, 1540 (1949); D. Saxon and J. Richards, Phys. Rev. **76**, 186 (1949); H. Agnew, Phys. Rev. **77**, 655 (1950); R. Caldwell, Phys. Rev. **78**, 407 (1950).

[18] The energies (see reference 19) of the most prominent K x-rays of Pr are: $K\alpha_2=35.6$ kev; $K\alpha_1=36.1$ kev; $K\beta_1=40.8$ kev, while the relative intensities are approximately (see reference 19) 3:6:2, respectively. Constructing Gaussian curves with suitable half-widths, and adding them, an approximate Gaussian results with its peak at 36.7 kev.

[19] A. H. Compton and S. K. Allison, reference 10, pp. 640–1, 785.

[20] West, Meyerhof, and Hofstadter, Phys. Rev. **81**, 141 (1951).

[21] F. K. McGowan, Phys. Rev. **85**, 151 (1952).

[22] D. W. Engelkemeir (private communication).

by others,[22,23] but does not seem to be characteristic of NaI(Tl), since other laboratories[24] have found NaI(Tl) spectrometers to be linear to quite low energies. The reasons for the different results are not clear.

Due to the limited resolving power of scintillation spectrometers in this low energy region, the overlapping peaks were most conveniently resolved by constructing individual components with "ideal" shapes. An examination of the Ce^{141} curve (Fig. 5b) showed the 146-kev gamma-ray and Pr K x-ray peaks to be almost pure Gaussian curves, with half-widths proportional to (pulse-height)$^{\frac{1}{2}}$. The 85-kev UY peak examined through a lead absorber (Fig. 5e) was similarly found to be reasonably "clean" Gaussian curve, with the expected half-width. The more complex curves were analyzed by constructing individual Gaussian curves of assumed energies and intensities and suitable half-widths.

While most of the UY gamma-rays reported here were observed as separate peaks, some of the ones listed in Table III were presumed to be present only as a result of analysis of the complex curves. Thus, there is little doubt concerning the existence of gamma-rays at 22, "61," 85, and 167 kev and with somewhat less certainty, at 107 kev; some of these lines may, of course, be composites. As mentioned below, there may also be unresolved K x-rays at about 93 kev. Using these energies and "ideal" curves with appropriate half-widths, it was not found possible to account for the shape of the experimental curves without assuming that another gamma-ray was present at 122 kev. Of course, the energy and intensity assumed for this component were quite arbitrary; the energy was chosen to fit the proposed decay scheme (Fig. 7). This component would then match a K-conversion line observed in the magnetic spectrometer at 9 kev. The tail at the high energy side of the 167-kev peak (Fig. 5f and 5g) was too wide to be accounted for by only one peak of Gaussian shape, and hence the assumption that both 210- and 230-kev gamma-rays were present. Here too, the choice of energies and intensities was somewhat arbitrary.

TABLE III. Experimental data on relative gamma-ray intensities.

Absorber	Gamma-ray energy	Absorption correction factor	Relative intensity
Al	22	1.46	1.7
0.2 g/cm²	85	1.00	1.00
Ta	59 } Av=61*	137.	0.40
1.29 g/cm²	63 }	65.4	
Pb			
0.56 g/cm²	85	3.0	0.70
Ag	85	81.	0.78
1.70 g/cm²	107	10.	0.06
	122	4.9	0.021
	167	2.2	0.016
	210	1.6	0.003
	230	1.5	0.001
Ag	85	850.	1.16
2.60 g/cm²	107	34.	0.075
	122	11.5	0.022
	167	3.3	0.019
	210	2.0	0.003
	230	1.9	0.001

* Assumed to have a 63/59 intensity ratio of 2.

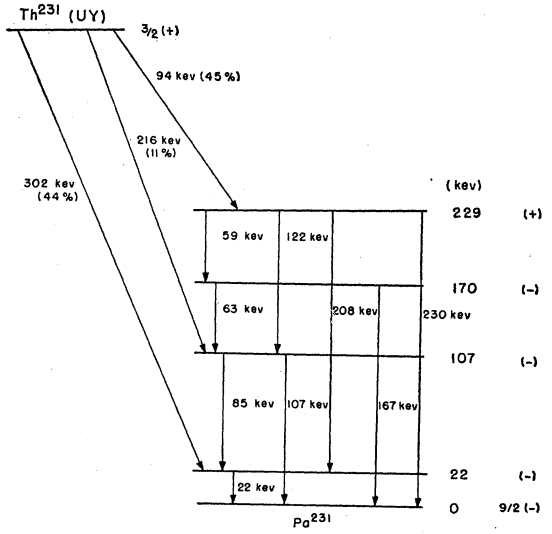

FIG. 7. Suggested decay scheme for Th^{231} (UY).

Although previous work[3] had indicated the presence of L x-rays, these were not resolved in the present measurements, and presumably appeared only as a widening of the 22-kev peak. Analysis of the peak shows that it must be, in the main, a gamma-ray of 22 kev (the energy uncertainty predominantly positive due to possible peak shift by L x-rays). The intensity of the gamma-ray itself is less than that indicated in Table IV; however, it seems unlikely that the L x-rays contribute more than about 30 percent.

The very high efficiency of the NaI(Tl) crystal for gamma-rays of low energy made possible semiquantitative estimates of relative intensities. Up to 200 kev, over 80 percent of the photons entering the crystal should have interacted with it. Up to 100 kev most of the interaction is photoelectric absorption and, were it not for the escape peak, the main peak intensity would fairly well measure relative intensity in the source. Even above 100 kev, the observed peak represents more than the photoelectric absorption, due to reabsorption of Compton-scattered photons in the crystal.[25] For example, Fig. 5b shows that at 146 kev, most of the gamma-ray pulses lie in the peak. At lower energies

[23] R. W. Pringle and S. Standil, Phys. Rev. **80**, 762 (1950).
[24] V. O. Eriksen and G. Jenssen, Phys. Rev. **85**, 150 (1952); West, Meyerhof, and Hofstadter, Phys. Rev. **81**, 141 (1951); Jentschke, Eby, Taylor, Remley, and Kruger, Phys. Rev. **83**, 170 (1951); **84**, 1034 (1951) (for electrons).

[25] J. A. McIntyre and R. Hofstadter, Phys. Rev. **78**, 617 (1950); R. Hofstadter and J. A. McIntyre, Phys. Rev. **80**, 631 (1950).

TABLE IV. Averaged values for relative gamma-ray intensities.

Gamma-ray energy	Relative intensity
22 (+L x-rays)	1.7
61 {59, 63}	0.40*
85	1.00
105	0.06_5
122	0.02
167	0.018
210	0.003
230	0.001

* May be too high.

the escaping I K x-ray reduces the efficiency but even at 60 kev the effect is no larger than 10 percent.[26]

The relative intensities of the gamma-rays were calculated by multiplying the peak heights by (pulse height)$^{\frac{1}{2}}$,[27] and by the appropriate absorption correction factor.[10,28] The accuracy of the latter correction factor was poor for some of the gamma-rays, due to the large attenuations used, with attendant accentuation of the errors in values of the absorption coefficients. In Tables III and IV are listed the observed relative intensities, after correction. Escape peak corrections or corrections for crystal stopping power were not made; the resulting errors were probably less than 20 percent. In most cases, the absorption correction was probably the most serious source of error. The abundance of the 61-kev component is quite uncertain. The absorption correction (Ta curve) is quite large, and its average magnitude depends upon the intensity division between the 59 and 63 kev components. The ratio (59:63 intensity) was assumed to be 1:2, in order to equalize the transition intensities (conversion electrons+gamma-rays). An examination of Fig. 5c, taken with no added absorber, would indicate the 61:85 ratio to be of the order of 1:10 rather than the 0.4 of Table III. This evidence, however is not reliable, since a relatively small shift in the construction of the 85-kev peak would increase the intensity available to the 61 kev component.

In addition to absorption correction errors, as mentioned above, the construction of the 122-, 230-, and 210-kev peaks involved some arbitrariness in choice of intensity. The uncertainty in the intensity of the 22-kev peak was increased by the presence of unresolved L x-rays. The intensity discrepancy (0.70:1.00) in the 85-kev lines observed with and without Pb absorber (Table III) may indicate the presence of an unresolved component between 85 and 107 kev, since the Pb absorber would attenuate such a component (K-edge $=88$ kev). K x-rays (93 kev) are known to be present to a small extent due to conversion of the 167- and 208-kev transitions and to a possibly larger extent due to 122-kev conversion [Table II, $K(5)$].

[26] F. K. McGowan (private communication).
[27] For a Gaussian curve, area $\propto$ peak height$\times$half-width.
[28] M. T. Jones, Phys. Rev. **50**, 110 (1936).

DISCUSSION

It did not appear possible to construct a decay scheme for UY consistent with all the experimental facts. The scheme we present (Fig. 7) is in the main a consequence of the observed energy balances between the major and most convincingly assigned gamma-rays, the 59, 63, and 85 kev, and the beta-energy differences. To these are added the well established (Fig. 5g) though less accurately measured 167-kev gamma and the 22-kev gamma (Fig. 5c and Table II).

As was discussed in the gamma-ray experimental section, the precise allocation of energies and relative intensities of the remaining gammas, 107, 122, 208, and 230 kev, was guided by the level structure of this decay scheme, though in each case the gamma-ray spectrometer results seemed to require the presence of more than one gamma in the energy range 90–130 and similarly for the range 200–240 kev. Supporting evidence for the existence of the 122-kev gamma is derived from the most prominent low energy conversion line $K(5)$; and no assignment for line $K(6)$ other than as the K line of a 208-kev gamma seems reasonable.

The absence of an L conversion line for the 122-kev gamma at *ca* 102 kev (present to <0.02 percent of total beta) implies an unacceptably large K/L conversion ratio; this must be regarded as one of the inconsistencies in the picture.

No concern need be given to the absence of an L conversion line for the 208-kev gamma as it is expected to be of lower intensity than the K line which is barely observable. As for the L line of the 167-kev gamma, it too may be expected to be weaker than the K line which constitutes only a fraction of the line $M_{II}(2)$, $K(7)$. These considerations are strengthened by reference to the low intensities of the unconverted gammas (Table IV).

The intensity of the gammas was related to the intensity of the betas and conversion electrons by normalizing the 85-kev gamma to match the sum of the intensities of the two weaker betas, correcting for the small branching by way of the 107-kev gamma. The one glaring inconsistency in the proposed scheme is associated with intensity considerations of the weak (94-kev) beta and the 59-kev—63-kev gamma-sequence. Referring to Fig. 7 and Tables I, II, and IV, it will be seen that, since the intensities of the 122-, 208-, and 230-kev transitions are low, the 59-kev gamma plus conversion electron intensities should be nearly equal to the intensity of the weak beta; and since the 167-kev transition is weak compared to the 63-kev transition, the latter should be comparable to the 59-kev total intensity. Therefore, it is reasonable to divide the observed gamma-intensity of the "61-kev" line between the 59- and 63-kev gammas so as to equate their total (gamma plus conversion electron) intensities. Using the most favorable (largest) value for the total intensity of the 61-kev gamma (40 percent of that of the 85-kev

gamma) and accounting for the conversion of the 85-kev transition, for the small branches by way of the 107-, 122-, and 167-kev gammas, and for the contribution of the 216-kev beta, one obtains the intensity figures summarized in Table II.

The necessary subdivision of the "61-kev" gamma into 59-kev and 63-kev components (1:2) is not in conflict with the fact that the gamma-peak was found at 61 kev, instead of *ca* 62 kev, in consideration of the accuracy of the gamma-measurements. The burden of the discrepancy is, in these calculations, placed entirely on the 59-kev and 63-kev transitions, which are seen to be too low by about a factor of 2, comparing the 50 percent intensity of the latter with the 80 percent figure for the sum of the 85-kev plus 167-kev transitions fed by the softest beta [11/(45+11)=20 percent contribution of the 216-kev beta subtracted off]. A reduction of the relative intensity of the 94-kev beta by a factor of *ca* 3 is, however, required if the intensities of the two low energy betas and the gammas of 59, 63, and 85 kev are to be brought into agreement. So great a reduction appears irreconcilable with the Kurie analysis (see discussion in experimental section). That the low energy beta is present in substantial abundance is also consistent with the experimental results of Stoker *et al.*[5] who found, with sources probably thicker than were used in the work reported here, betas of energy 390, 190, and 100 kev in relative intensities 1:2:2.

Based on the experimental beta branching ratios, the logft values for the beta-transitions are 6.2 (302 kev), 6.0 (216 kev), and 4.3 (94 kev). The low value for logft for the 94-kev beta is exceptional for such high Z; the only other value below 5.0 for $Z \geq 74$ is that for Pb^{214} (4.70). This may be taken as further evidence that the experimental value for the intensity of the 94-kev beta may be too high; a threefold reduction in the intensity would result in a logft=4.9.

It is also possible that the low intensity of the 59 kev–63 kev sequence is due to a higher intensity of the 122-kev cross-over transition than observed. Analysis of the 85-kev gamma-ray line (Figs. 5c, e, and Table III) indicates a maximum possible (though unlikely) intensity of K x-ray (*ca* 93 kev) of about 30 percent of that of the "85-kev peak." This figure would be sufficient to account for most of the intensity misfit. However, so large a K conversion coefficient would aggravate the already too high K/L conversion ratio limit discussed above. Also the intensity of K electrons would then exceed the *ca* 13 percent figure set as a rough upper limit (Table II) for $K(5)$.

There is only a slight broadening of the 22-kev gamma-peak, (Fig. 5c), insufficient to resolve any L x-rays. If an average value for the fluorescent yield of all L vacancies of 40 percent is assumed[29] a conversion coefficient of unity for the 22-kev transition is required to match the observed intensity of the 22-kev gamma (plus its conversion electrons, of which only the relatively weak L_{III} and higher energy lines were observed) to the sum of the beta-intensities.

Sufficient evidence is available to allow plausible guesses as to spin and parity assignments of the levels of the Th^{231} and Pa^{231} nuclei. However, such arguments depend intimately on the validity of the decay scheme assumed, and this is considered to be too uncertainly established to warrant extensive discussion. In Fig. 7 we indicate spins and parities for the levels consistent with the ft values and transition probabilities observed, and with the predictions of the strong spin-orbit coupling nuclear shell model.[30]

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[29] B. B. Kinsey, Can. J. Research **26A**, 404 (1948).
[30] M. G. Mayer, Phys. Rev. **78**, 16 (1950).