PHYSICAL REVIEW VOLUME 99, NUMBER 1 JULY 1, 1955

Decay Properties of U^{232}†

FRANK ASARO AND I. PERLMAN
Radiation Laboratory and Department of Chemistry, University of California, Berkeley, California

(Received March 16, 1955)

The alpha and gamma spectra of U^{232} have been studied with an alpha-particle spectrograph and gamma-ray scintillation, proportional, and coincidence counters. Alpha groups of 5.318 Mev (68 percent), 5.261 Mev (32 percent) and 5.134 Mev (0.32 percent) and gamma rays of 57.9 kev (0.21 percent), 131 kev (0.075 percent), 268 kev (0.004 percent), and 326 kev (0.004 percent) were observed. The half-life of the 58-kev first excited state of Th^{228} was found to be less than 10 microseconds. Spins and parities are assigned to the energy levels, and the results are evaluated with respect to the developing theory and systematics of complex alpha spectra and excited states of even-even nuclei.

INTRODUCTION

THE uranium isotope, U^{232}, is a beta-stable alpha emitter with a half-life of 73.6 years[1] and was first identified following its growth from the shorter-lived β^--emitter Pa^{232} which had been prepared by the $(d,2n)$ reaction on Th^{232}.[2] The alpha-particle energy has been determined by range measurement as 5.31 Mev[3] and 5.27 Mev.[4] It could be inferred that the U^{232} alpha spectrum was complex because prominent conversion electrons of an ~60-kev gamma ray were observed in coincidence with alpha tracks in a photographic emulsion.[5] An early part of the present study, in which there was found an alpha group of 58-kev lower energy than the main group, has already been reported.[6]

There has been added recent interest in the alpha spectra of even-even nuclei because they display prominently series of energy levels which have been interpreted as rotational states.[7–9] The theory of Bohr and Mottelson which explains these rotational bands as a consequence of collective motions in highly deformed nuclei predicts a simple relationship of the energy spacing between members of the band: $E_{\text{rot}} \propto I(I+1)$, where I refers to the spin which in this case is restricted to even integral numbers. As will be mentioned further in the Discussion, this simple expression applies to a limiting condition in which the rotational frequency is sufficiently slow to allow the nuclear structure to adjust adiabatically to the changing electric field. Where this situation does not apply, the rotational levels are perturbed and correction terms must be applied. It is noted that the heaviest nuclei show "pure" rotational bands and as the closed-shell region around lead is approached the perturbation becomes more and more severe. In this context, it will be seen that Th^{228} (alpha decay of U^{232}) shows a just discernible departure from the ideal case and therefore represents the entry into the "light-element" or closed-shell region.

Another aspect of note in the decay of U^{232} is the appearance of a low-lying level which does not belong to the rotational band. This particular state with spin 1, odd parity, occurs in a limited region (around 136 neutrons). It was the subject of another publication[10] and will be discussed further below.

METHODS

Preparation of source.—The U^{232} sources for the present study were prepared in two ways. In one of these, Th^{232} was bombarded with protons resulting in U^{232} by the following reactions[11]:

$$Th^{232}(p,n)Pa^{232} \xrightarrow{\beta^-} U^{232}.$$

The other method involved intensive neutron irradiation of ionium (Th^{230}):

$$Th^{230}(n,\gamma)Th^{231} \xrightarrow{\beta^-} Pa^{231}(n,\gamma)Pa^{232} \xrightarrow{\beta^-} U^{232}.$$

Inasmuch as Th^{232} was present in the irradiated ionium, U^{233} was also made by a first-order neutron capture reaction. However, its contribution to the radioactivity was quite insignificant; the alpha spectrum showed a possible peak of the U^{233} position of maximum intensity 0.06 percent relative to the U^{232} groups. The factors which produced this favorable ratio were: (1) the ionium had been considerably enriched by electromagnetic separation[12] from the initial source materials, (2) the

---

† This work was performed under the auspices of the U. S. Atomic Energy Commission.

[1] Sellers, Stevens, and Studier, Phys. Rev. **94**, 952 (1954).

[2] J. W. Gofman and G. T. Seaborg, The Transuranium Elements: Research Papers (McGraw-Hill Book Company, Inc., New York, 1949), Paper No. 19.14, National Nuclear Energy Series, Plutonium Project Record, Vol. **14B**, Div. IV., p. 1427.

[3] A. H. Jaffey (unpublished data, 1948).

[4] H. Kahn and G. A. Linenberger, reported in Los Alamos Scientific Laboratory Report LAMS-151, October, 1944 (unpublished).

[5] D. C. Dunlavey and G. T. Seaborg, Phys. Rev. **87**, 165 (1952).

[6] Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).

[7] A. Bohr and B. R. Mottelson, Phys. Rev. **89**, 316 (1953); **90**, 717 (1953); Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **27**, No. 16 (1953).

[8] A. Bohr, *Rotational States of Atomic Nuclei* (Ejnar Munksgaard, Copenhagen, 1954).

[9] F. Asaro and I. Perlman, Phys. Rev. **91**, 763 (1953).

[10] Stephens, Asaro, and Perlman, Phys. Rev. **96**, 1568 (1954).

[11] We wish to thank Dr. Louis M. Slater for making this sample available to us.

[12] For typical available compositions of enriched ionium, see: Harmatz, McCurdy, and Case, Oak Ridge National Laboratory Report ORNL-1724, 1954 (unpublished). We wish to thank

TABLE I. Alpha-particle spectrograph exposures.

Exposure number	Source of U^{232}	Slit width of spectrograph source (inch)	Duration of exposure	Activity of sample (dis/minute)	Time after Th^{228} removal
134	$Th^{232}(p,n)Pa^{232} \xrightarrow{\beta^-} U^{232}$	$\frac{1}{8}\times 1$	46 hr	10^5	~1 week
245	$Io^{230}(n,\gamma)Th^{231} \xrightarrow{\beta^-} Pa^{231}$ $Pa^{231}(n,\gamma)Pa^{232} \xrightarrow{\beta^-} U^{232}$	$\frac{1}{8}\times 1$	14 hr	6×10^8	~1 month
277	Same sample as used in exposure 245	$0.018\times\frac{3}{4}$	46 hr	...	7 months
278	Same sample as used in exposure 245	$0.018\times\frac{3}{4}$	47 min	...	7 months

neutron capture cross sections[13] for Th^{230} and Pa^{231} are much larger than that for Th^{232}, (3) the half-life for U^{233} is long compared with U^{232}, and (4) the uranium was separated before all of the intermediate Pa^{233} had decayed to U^{233}.

Alpha-particle spectra.—All samples for the alpha-particle spectrograph were prepared by vacuum sublimation of the chloride onto a platinum plate which was masked to present a band 1 inch×⅛ inch. This sample mounting technique, as well as the equipment and methods for taking alpha spectra, have been described in earlier reports.[14–16] As before, the alpha particles were caught on a photographic plate and the track count was plotted according to position on the plate.

Gamma-ray measurement.—Gamma-ray spectra were measured for the most part with a sodium iodide scintillation counter coupled to a 50-channel pulse-height analyzer. In some experiments a xenon-filled proportional counter was used to produce the pulse for the analyzer.

Coincidence counting methods used in this and similar studies have been described elsewhere.[10,17] Briefly, a gamma-ray pulse is fed into a 50-channel pulse-height analyzer which registers only when triggered through a gate circuit. For alpha-gamma coincidences the pulse produced by a zinc sulfide screen in optical contact with a photomultiplier tube is used to open the gate; for gamma-gamma coincidences, another sodium iodide crystal counter was used.

RESULTS

Alpha Spectrum

Four exposures, involving two U^{232} preparations, were made and the histories are given in Table I. The first measurement (Exp. 134) was made with a relatively weak sample before more active preparations had been made. Numbers 245 and 277 were moderately long exposures of an intense source aimed principally at finding low-intensity alpha groups. The more intense alpha groups register too many tracks for convenient counting. A shorter exposure (Exp. 278) was then made to get more accurate data on the abundances and energies of the most prominent groups.

As a result of these several experiments, three alpha groups were found for U^{232} as summarized in Table II. Two of these are the characteristic high-intensity groups of an even-even alpha emitter leading to the ground state and first excited state of the product. The energy of the main group was found to be 5.318 ±0.002 Mev and the second group was lower in energy by 57.2±1.2 kev. (The 5.421-Mev [18] alpha group of Th^{228}, which grew into the U^{232} sample, was used as the energy standard for these measurements.) When the differences in recoil energies from the two alpha groups are included, the first excited state is found to lie at 58.2 kev. The intensity of α_{58} was found to be 32 percent.[19]

Besides the two principal groups a weak alpha group

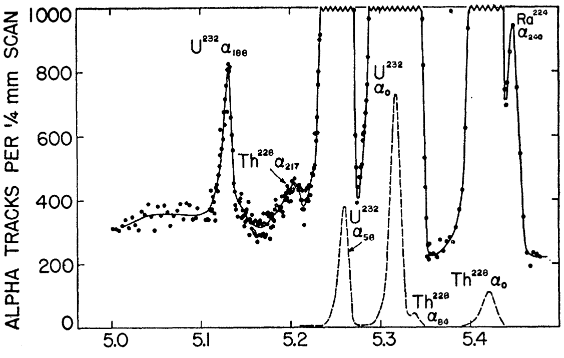

FIG. 1. U^{232} alpha spectrum with daughters partially grown in. —— Exposure 277. ------- Exposure 278 (ordinate scale should be multiplied by 2).

---

members of the Electronuclear Research Division of Oak Ridge National Laboratory for making some enriched ionium available to us.

[13] See: *Neutron Cross-Sections*, U. S. Atomic Energy Commission Report AECU-2040 (Technical Information Division, Department of Commerce, Washington, D. C., 1952).

[14] F. L. Reynolds, Rev. Sci. Instr. **22**, 749 (1951).

[15] Asaro, Reynolds, and Perlman, Phys. Rev. **87**, 277 (1952).

[16] Asaro, Thompson, and Perlman, Phys. Rev. **92**, 694 (1953).

[17] Asaro, Stephens, Thompson, and Perlman, Phys. Rev. **98**, 19 (1955).

[18] Asaro, Stephens, and Perlman, Phys. Rev. **92**, 1495 (1953).

[19] α_{58} refers to the alpha group leading to the 58-kev state.

TABLE II. Results of spectrograph exposures.

Exposure number	α particle energy separation between α_0 and α_{58} (kev)	Separation between α_0 and α_{58} including difference in nuclear recoil (kev)	Intensity of α_{58} relative to total U^{232} α particles (%)	α decay energy separation between α_{58} and α_{188} (kev)	α decay energy separation between α_0 and α_{188} (kev)	Intensity of α_{188} relative to total U^{232} α particles (%)	α particle energy of U^{232} α_0 relative to Th^{228} α_0 (5.421 Mev) (Mev)	α particle energy of U^{232} α_{58} relative to Th^{228} α_0 (5.421 Mev) (Mev)
134	58±2	$59.{}_5$	31	...	...	...	...	...
245	55±4	56	35	...	189	0.3	5.317	...
277	...	...	...	$129.{}_5$	...	0.32±0.03	...	5.260±2
278	57.2±1.2	58.2	32±1	...	...	...	5.318±2	5.261±2
Best value	57.2	58.2	32		188	0.32	5.318	

of 0.32 percent abundance was identified as belonging to U^{232}. This group leads to a state 188 kev above the ground state. Gamma rays originating from this state and others will be described below. No other alpha groups belonging to U^{232} were noted but the limits of detection varied with the proximity to the observed groups. Over the energy range 5.07 Mev→4.83 Mev an upper limit of 0.01 percent could be set. At 4.822 Mev there was a small peak (0.06 percent of the U^{232} intensity) which is probably due to U^{233} as the energy agreement is good and it is expected that there would be some present.

The alpha spectrum of U^{232} with some of its daughter activities is shown in Fig. 1. The solid line curve was obtained from Exp. 277 in which the principal peaks were too strong to be counted. The positions and intensities of the strong groups taken from Exp. 278 are shown as the broken line curve of Fig. 1.

Gamma Rays and Decay Scheme

The partial decay scheme for U^{232} based on the present studies is shown in Fig. 2. It will be noted that a third excited level in Th^{228} is indicated by gamma radiation although the alpha group populating this state could not be seen. The experiments will be discussed in terms of the successive energy levels. Some of the gamma rays measured were extremely weak in intensity and a number of measurements of the gamma spectrum were made after repeated and varied chemical purification steps to make certain that these radiations did not arise from Th^{228} decay products. The only nonseparable substance which could have been present was U^{233} and an upper limit to its intensity could be set on the basis of the alpha spectrum.

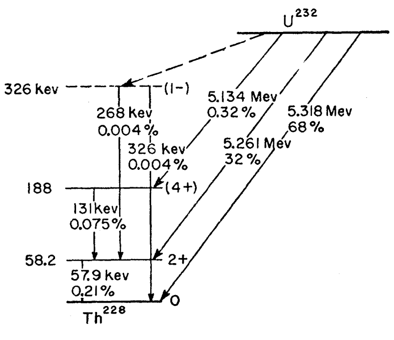

FIG. 2. U^{232} decay scheme.

The measurements on the photons included: (1) identification and intensity measurements with a scintillation spectrometer, (2) alpha-gamma and gamma-gamma coincidence determinations, (3) xenon-filled proportional counter measurements of energy. In all, 13 sets of measurements were made employing six different repurified samples of U^{232} from a common original stock.

The 58-kev state.—As indicated by the alpha spectrum, the first excited state lies at 58.2±1.2 kev and is populated directly to the extent of 32 percent. Further possible population of this state by transitions from higher states cannot be much in excess of 0.32 percent.

Several measurements with the scintillation spectrometer in which the 59.6-kev gamma ray of Am^{241} was used as an energy standard showed a peak at 57 to 58 kev. Two measurements with a xenon-filled proportional counter gave the best energy determination as 57.9±0.3 kev. The prominent Am^{241} photon also served as an intensity standard. The best determination of the intensity was 2.1×10^{-3} relative to total alpha disintegrations. The total conversion coefficient is therefore 152 which agrees well with the value 120 for the *L*-shell conversion coefficient for an *E*2 transition which was estimated from available theoretical data.[20,21] This assignment confirms the expected 2+ designation for the first excited state of Th^{228}.

This same state has been observed by others[22–24] in the beta spectrum of $MsTh_2(Ac^{228})$. Brodie[23] gave the energy of the transition as 57.0 kev and Kyles, Campbell, and Henderson[24] reported 56.75 kev. These two reports concluded that the transition is *E*2 although the conversion coefficients differed somewhat from those deduced here.

A puzzling feature of the 58-kev transition is an apparent discrepancy concerning the lifetime of the 2+

---

[20] Rose, Goertzel, and Swift, *Tables of Conversion Coefficients* (privately circulated).

[21] Gellman, Griffith, and Stanley, Phys. Rev. **85**, 944 (1952).

[22] D. H. Black, Proc. Roy. Soc. (London) **A106**, 632 (1944).

[23] W. D. Brodie, Proc. Phys. Soc. (London) **A67**, 265 (1954).

[24] Kyles, Campbell, and Henderson, Proc. Phys. Soc. (London) **A66**, 519 (1953).

state. Two groups[24,25] working on the beta spectrum of MsTh_2 have reported the lifetime of this state to be greater than 10 milliseconds. A third group has reported the lifetime to be larger than 0.5 second.[26] From the present results it is deduced that the lifetime of this state is less than 10 microseconds. In two measurements the coincidence rate between alpha particles and 58-kev gamma rays was 0.16 percent and 0.18 percent per alpha particle. These values correspond within experimental error with the above-mentioned intensity of 0.21 percent for the 58-kev photon, showing that it is in coincidence with the alpha particles within the resolving time of the coincidence circuit which was about 10 microseconds. In addition coincidences were observed between the 130-kev gamma ray and L x-rays. These L x-rays arise mainly from the conversion of the 58-kev gamma ray.

It is probable that the methods[24–26] by which delays between the events were noted could not distinguish between a long delay and a very rapid coincidence. It is to be expected[7,27] that, in a region such as this well away from closed shells, the $E2$ transitions from the first excited state would be abnormally rapid rather than slow.

The 188-kev state.—A state at this energy is defined uniquely by the existence of a U^{232} alpha group with an energy lower than the ground state transition by the appropriate amount. As already mentioned, this state is populated to the extent of about 0.32 percent.

The only gamma-ray transition observed in the present study which depopulates this state is one of 131 kev. It leads from the 188-kev state to the 58-kev state as determined by gamma-gamma coincidence measurements between 131- and 58-kev photons and between the 131-kev gamma ray and L x-rays. This situation is similar to that of other even-even nuclei among the heavy elements in which a series of states exist which are believed to belong to a rotational band and follow the sequence $0+$, $2+$, $4+$, $\cdots$. In none of these cases has a crossover transition been seen from the second even spin state to the ground state. It remains now to be seen if some of the other criteria for this sequence exist.

One of the properties of a pure rotational spectrum is that the energy spacings are proportional to $I(I+1)$, where I is the spin. The ratio of energies between the spin 4 and spin 2 states should be accordingly 20/6 or 3.33. In the present situation the ratio is 188/58 or 3.24.

If the 131-kev transition takes place between $4+$ and $2+$ states it must be of pure $E2$ character. Two measurements were made of the alpha-gamma coincidence rate for the 131-kev gamma ray. These gave intensities for the gamma ray of 0.05 and 0.06 percent. Two more precise measurements of the gamma spectrum gave a value of 0.075 percent for the best intensity of the 131-kev gamma ray. Inasmuch as the 188-kev state is populated to the extent of 0.32 percent the conversion coefficient becomes ~ 3.2. This is in good agreement with the expectations for an $E2$ transition, it is definitely too high for an $E1$ transition, and is about a factor of 3 too low for an $M1$ transition. There is some radiation observed at about 85 kev (see Fig. 3.) Although it may include scattered radiation from the 131-kev gamma ray, its abundance represents the maximum intensity of K x-rays in the decay of U^{232}. This abundance is 0.4 of the abundance of the 131-kev gamma ray, giving a maximum K conversion coefficient of 0.4. This is in good agreement with the expectations for an $E2$ transition but is a factor of 20 too low for an $M1$ transition.

A gamma-ray transition of about this energy (128 kev) was studied by Brodie[23] and by Kyles, Campbell, and Henderson[24] in the conversion electron spectrum from MsTh_2. As pointed out[24] the L subshell conversion ratios and the low limit set on the K conversion indicates an $E2$ transition, with which our conclusions agree. The same gamma ray (129 kev) was seen by Box and Klaiber[28] using a scintillation spectrometer.

These workers[22,24] also studied a transition of about 184 kev which they interpreted as an $M1$ transition and in their decay scheme was placed as the crossover transition from a level of this energy to the ground state for which the cascading transitions are those of 128 kev and 58 kev. In order to fit spins and parities to these conditions the sequence for the ground state and next two higher states was postulated[24] to be $0+$, $2+$, $1+$. There are two reasons why a state at 184 kev which meets this condition cannot be the same as our 188-kev state defined by the alpha decay of U^{232}. In the first place an even-even alpha emitter cannot populate directly a state of odd spin and even parity. The other argument has to do with intensities of gamma rays. According to the data and deductions of Brodie[23] and of Kyles and co-workers,[24] the intensities of the 128-kev and 184-kev photons should be about the same. From our measurements the intensity of the 131-kev gamma ray is at least 30 times greater than a peak at 184 kev (see Fig. 3). In order to reconcile these results it is necessary to say that either there is a $1+$ state at

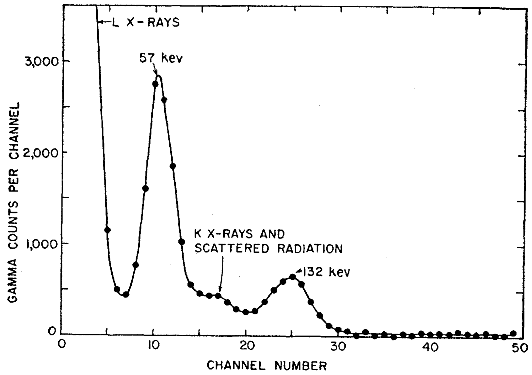

FIG. 3. U^{232} low-energy gamma spectrum.

---

[25] Lecoin, Perey, and Teillac, J. phys. radium **10**, 33 (1949).

[26] F. Suzor and G. Charpak, J. phys. radium **15**, 682 (1954).

[27] M. Goldhaber and A. W. Sunyar, Phys. Rev. **83**, 906 (1951).

[28] H. C. Box and G. S. Klaiber, Phys. Rev. **95**, 1247 (1954).

184-kev (populated from the beta decay of MsTh$_2$) which is different from our 188-kev state (from alpha decay of U^{232}) or the observed 184-kev transition involves higher-lying states of Th^{228} which also would not be expected to be seen in alpha decay processes. An added datum which must be considered in the over-all interpretation is the failure of Box and Klaiber[28] to observe a 184-kev photon in their scintillation counter study of MsTh$_2$. This could imply a high conversion coefficient for the 184-kev transition although it is rather difficult to set limits on the basis of their reported data.

The 326-kev state.—From the present work a level at this energy (see Fig. 2) could only be inferred from gamma-ray measurements, as the alpha-particle intensity is apparently too weak for direct detection. The evidence for the state is based on the observation of two gamma rays of 326 and 268 kev (see Fig. 4). The energy differences alone suggest transitions from a 326-kev level to the ground state and the 58-kev state, respectively.

The intensities of the two peaks as determined with the scintillation spectrometer were both about the same and amounted to about 4×10^{-3} percent of the total alpha disintegrations for each. An alpha-gamma coincidence measurement for the 268-kev gamma ray gave an intensity of 5×10^{-3} percent. Similarly, if L x-rays were used to gate the scintillation spectrometer it was found that the 268-kev gamma rays were in coincidence and the intensity came out to be 3×10^{-3} percent per alpha particle. Much less abundant coincidences with L x-rays were found for the 326-kev gamma ray. Because the L x-rays arise predominantly from the 58-kev transition it is probable that the 268-kev gamma ray goes to the 58-kev state and the 326-kev gamma ray does not. The L x-ray coincidences with the 326-kev gamma ray could be due to impurities or to the decay of still higher excited states populated by U^{232} alpha emission.

These intensity, coincidence, and energy considerations imply that a single state at 326 kev is responsible for both of these photons as shown in Fig. 2. Box and Klaiber[28] also found a pair of gamma rays differing by 58 kev and having approximately the energies seen here. Their values were 336 and 278 kev. Furthermore, they found coincidences between both of these and a gamma ray of 790 kev, which is also consistent with the supposition that both originate from a common level. However, the spectrum shown by these authors would indicate that the 336-kev gamma ray is at least twice as intense as the 278-kev gamma ray while in our measurements the 278-kev photon seemed to be somewhat more intense. This may suggest that there are two gamma rays of about 336 kev, one of which occupies the position as shown by us (and by Box and Klaiber) and the other is a transition between higher levels. Significantly, Kyles and co-workers[24] report conversion lines from a gamma ray of 336 kev but not from one at 278 kev.

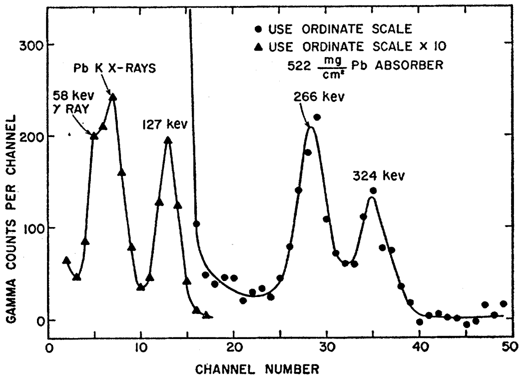

FIG. 4. U^{232} high-energy gamma spectrum.

In the absence of conversion coefficient data for our 326- and 268-kev transitions it is not possible to characterize them with certainty. We have been led to assign the 326-kev level $1-$ and consequently the transitions $E1$ by analogy with the well-characterized $1-$ levels found from other even-even alpha emitters in this region.[10]

DISCUSSION AND SUMMARY

The decay scheme for U^{232} shown in Fig. 2 bears a strong resemblance to those of other even-even alpha emitters in this region. In each case a rotational band consisting of $0+$, $2+$, and $4+$ states is seen. In some cases the $1-$ state lies between the $2+$ and $4+$ states but for U^{232} decay it lies above the $4+$ state. Just as in other cases, two $E2$ transitions are seen which cascade from the $4+$ to the $2+$ and $0+$ (ground state). The $1-$ state is de-excited by competing $E1$ transitions to the $2+$ and $0+$ states. To be sure, further information must be obtained to get independent evidence that the transitions in question for U^{232} are indeed $E1$.

As has already been discussed, there is a partial correlation of the energy levels found in the present study from the alpha decay of U^{232} and those from the beta decay of MsTh$_2$ found by others. There are also some points of apparent disagreement. Because of the extreme complexity of the MsTh$_2$ spectrum it is not possible to resolve these difficulties readily from existing data. Some of the information worth seeking in future studies on MsTh$_2$ has been mentioned in earlier discussion.