Decay of U^{232} (74 yr)*

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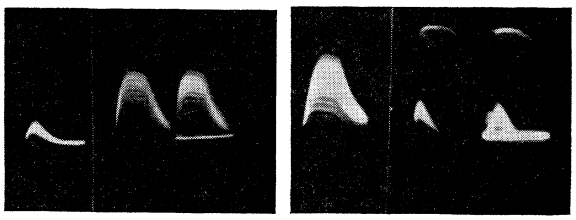
The decay of the α emitter U^{232} (74 yr) was studied by means of γ-ray scintillation counters and by a technique involving the impregnation of a scintillating crystal with this source. Excited states were found at 60 kev, 190 kev, and 330 kev. The first two are interpreted as rotational states of character 2+ and 4+ and the third as probably a 1− state. The branching ratios for transitions from the 330-kev state to the 2+ state and ground state are 57 percent and 43 percent respectively. The half-life of the 60-kev state was found to be shorter than 2×10^{-8} sec, in contrast to earlier reports based on a study of the β^- decay of Ac^{228}.

A CAREFUL study of the complex disintegration scheme of $Ac^{228}(MsTh_2)$ by Kyles, Campbell, and Henderson[1] made it appear plausible that the first excited state of Th^{228} lies at 57 kev. A study of coincidences between γ rays and conversion electrons from the highly converted 57-kev transition seemed to indicate that the 57-kev state has a half-life $\tau>10^{-7}$ sec, and a study of the distribution of electron pulses in time led the authors to conclude that $\tau>0.01$ sec.

The relative conversion coefficients for the 57 kev transition in the $L_{\rm I}$, $L_{\rm II}$, and $L_{\rm III}$ shells determined by Brodie[2] identify this transition as $E2$ if interpreted on the basis of Rose's[3] recent results on L conversion coefficients. This spin and parity assignment and also the energy fit well into the pattern of first excited states of even-even nuclei.[4] However, an $E2$ transition of this energy is expected to be faster even than a one-particle transition ($\tau<10^{-8}$ sec).

Recently Dunlavey and Seaborg[5] reported that a highly converted γ ray of about 60 kev follows the α emission of U^{232} in 31 percent of all disintegrations. It seems probable that this transition is identical with that following the β decay of Ac^{228}, and therefore we chose U^{232}, with its presumably very simple disintegration scheme, to search for the half-life of the first excited state of Th^{228}. As a first step we incorporated, by a method previously described,[6] a small amount of the U^{232} source[7] (kindly supplied by Dr. M. H. Studier of the Argonne National Laboratory) into a NaI(Tl) crystal which was then attached to a photomultiplier tube. If the 60-kev transition had a half-life $\tau>1$ μsec, which is the resolving time of the crystal, we would have observed pulses corresponding to conversion electrons from this transition, in addition to the α-ray pulses. No such pulses were observed, implying that $\tau<1$ μsec. We also prepared a U^{232} source mounted on 1 mg/cm^2 pliofilm and studied coincidences between two scintillation counters with thin anthracene crystals as detectors. The conversion electrons impinged directly on the crystal whereas the α particles had to traverse the pliofilm backing. Coincidences were observed between α particles and conversion electrons (a) by triggering an oscilloscope with α-particle pulses and observing pulses due to the conversion electrons from the 57-kev transition on the oscilloscope screen and (b) by triggering with conversion electron pulses and observing the α-particle pulses. In a similar fashion coincidences were observed between α particles and L x-rays (Fig. 1) and α particles and the unconverted 60-kev gamma ray by using a NaI(Tl) crystal as the photon detector. Since the oscilloscope "writing speed" was 1 μsec/inch, the above data indicated that the half-life of the 1st excited state of Th^{228} was $<\sim0.1$ μsec. Coincidence measurements between α particles and electrons were repeated using an oscilloscope with a "writing speed" of 0.2 μsec/inch and again the triggering pulses and the signal pulses of the oscilloscope appeared in prompt coincidence, hence $\tau<2\times10^{-8}$ sec, in agreement with expectations. As an additional check, a source of Ac^{228} was separated from its parent Ra^{228}



(a) (b)

FIG. 1. $\alpha-L$ x-ray coincidences in U^{232}. (a) The first pulse reading from left to right is the L x-ray pulse from detector 1 with which the oscilloscope is triggered during the coincidence run. The second pulse is self-triggered and shows α-particle pulses in detector 2. The third pulse is the α-particle pulse in detector 2 shown while the oscilloscope was being triggered by the L x-ray pulse of detector 1. (b) The roles of the α and L x-ray pulses were reversed and again the first pulse shows the triggering pulse (α particle in detector 2), the second pulse shows the L x-rays in detector 1, self-triggered, and the third pulse shows coincidences between the triggering pulse and the signal (L x-ray in detector 1).

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

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

[3] Rose, Goertzel, and Swift (private communication).

[4] G. Scharff-Goldhaber, Phys. Rev. **90**, 587 (1953); P. Stähelin and P. Preiswerk, Nuovo cimento **10**, 1149 (1953).

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

[6] Scharff-Goldhaber, der Mateosian, Goldhaber, Johnson, and McKeown, Phys. Rev. **83**, 480 (1951).

[7] The U^{232} sources were separated from their decay products immediately before use. The samples contained, however, a 30-fold excess (in weight) of U^{233}.

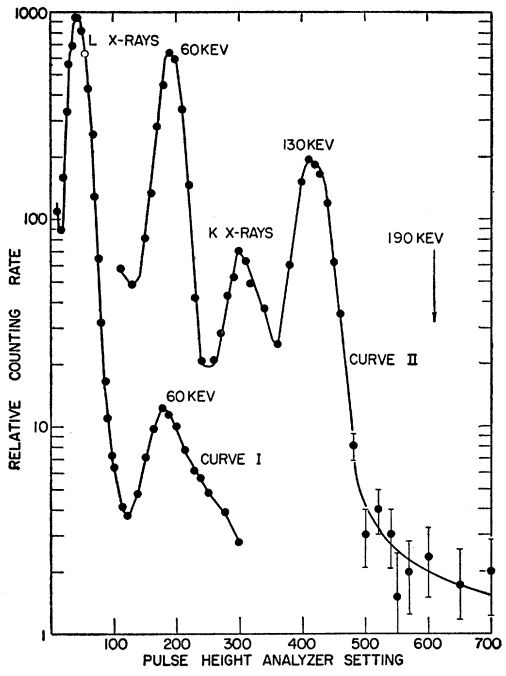

FIG. 2. Photon spectrum of U^{232} ($E \leq 200$ kev). Curve I, taken with a 1 cm^3 NaI(Tl) crystal with a Be window, shows the relative intensities of the 60-kev gamma rays and L x-rays. Curve II was taken with a (3 cm)^3 NaI(Tl) crystal. It shows the relative intensities of the 60-kev and 130-kev gamma rays and the K x-rays. The bump on the right side of the 60-kev peak in curve I was due to Bi K x-rays accompanying the decay of Pb^{212}, a daughter product of U^{232}. This residue of Pb^{212} was removed from the source by a repurification before Curve II was taken.

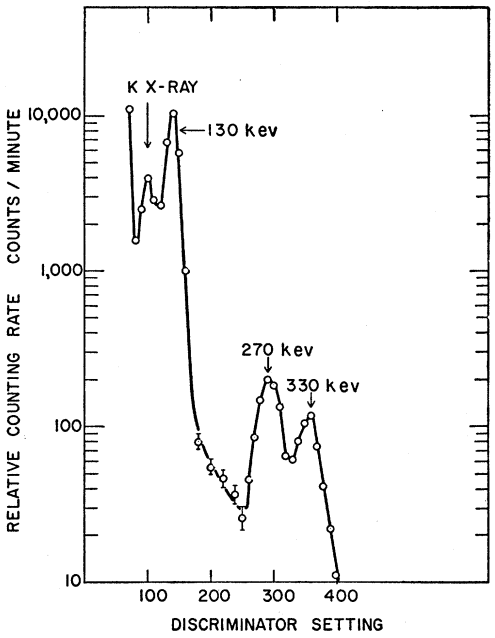

FIG. 3. Photon spectrum of U^{232} ($E \leq 400$ kev). Two well-resolved gamma rays are seen at 270 kev and 330 kev in addition to K x-rays and the 130-kev gamma ray. This run was made with the (3 cm)^3 NaI(Tl) crystal and a source of U^{232} which had been repurified to reduce the concentration of daughter products in the sample. The 159-, 239-, and 320-kev gamma-ray lines of Te^{123}, Pb^{212}, and Cr^{51}, respectively, were used to calibrate the energy scale.

and coincidences were observed between β particles and L x-rays; from this experiment we obtained an upper limit of 0.2 μsec for the lifetime of any electromagnetic transition strongly converted in the L shell. Since most of the L x-rays are due to the conversion of the 57-kev transition, this supports the results obtained with U^{232}.

The complete photon spectrum of the U^{232} source was investigated with a NaI(Tl) scintillation counter equipped with a single-channel pulse-height analyzer (Figs. 2 and 3). In the low-energy region unconverted γ rays of 60 ± 3 kev were seen as well as strong L x-rays, some K x-rays, and 130-kev gamma rays (Fig. 2). Assuming that practically all of the L x-rays are due to the conversion of the 60-kev transition, and applying a factor of 0.50 for the L-shell fluorescence yield,[8] we obtain a value of 120 for the L conversion coefficient of the 60-kev transition. We further find for the K conversion coefficient of the 130-kev transition a value of

TABLE I. Internal conversion coefficients and transition probabilities of γ rays from U^{232}.

Energy of γ rays (kev)	Assumed character of transition	Obs. rel. intensities of unconverted γ rays	Conversion coefficients[a] ϵ_K	ϵ_L	ϵ_M	ϵ_{tot}	Computed rel. int. of transitions	No. of transitions per 100 α's
60	$E2$	*1.00*	···	97 120[c]	32[b]	129	130	32
130	$E2$	0.51	0.36 0.38[c]	2.40	0.8[b]	3.56	2.3	0.57[d]
270	$E1$	0.037	0.035	0.005	···	0.04	0.039	0.0096
330	$E1$	0.028	0.022	0.003	···	0.025	0.029	0.0070

[a] Extrapolations of data in tables privately circulated by Rose *et al.* (reference 3).
[b] ϵ_M was estimated to be $\frac{1}{3}$ of ϵ_L. This agrees with measurements made by Brodie (reference 2).
[c] Experimentally determined values. See text.
[d] After these experimental results had been obtained, it was learned from a report by Rasmussen (reference 13) that Asaro, Stephens, and Perlman (private communication), by investigating the α spectrum of U^{232}, had also observed a 190-kev level. The α branch leading to this level was found to be 0.3 per 100 α's, in fair agreement with our result.

[8] B. B. Kinsey, Can. J. Phys. **A26**, 404 (1948).

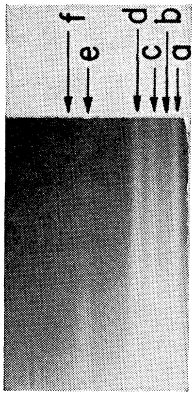

FIG. 4. Spectrum of photons coinciding with 60-kev γ rays of U^{232} as displayed with a gray wedge analyzer. This spectrum indicates that both the 330- and 270-kev γ transitions arise from a 330-kev level. Prominent lines are seen at ~35 kev (a), ~90 kev (K x-rays) (c), 130 kev (d), and 270 kev (e); f indicates the position of the 330-kev line which is missing in the coincidence spectrum. The position of the triggering 60-kev γ rays is given by b. The ~35-kev line may be the 42-kev gamma ray of U^{233} which coincides with a 56-kev gamma ray.

0.38 after correcting for crystal efficiency and fluorescence yield. Both conversion coefficients are compatible with $E2$ type transitions (see Table I), suggesting that Th^{228} may have a second rotational state (4+) at 190 kev. In this case $E_2/E_1=3.16$, which agrees within limits of error with 3.33 as derived from the Bohr-Mottelson $I(I+1)$ rule. Coincidence studies proved that the two γ rays are indeed in cascade. No cross-over gamma ray of 190-kev energy was seen; had it been present to the extent of 1 percent of the unconverted 130-kev transitions, it would have been detected.

The ratio of the unconverted gamma rays $N_{\gamma 130}:N_{\gamma 60} \approx 1:2$. If one uses the theoretical conversion coefficients for $E2$ transitions and the latest value[9] for the percentage of α transitions going to the ground state (68 percent), one finds that per 100 α transitions there are 32 transitions of 60 kev and 0.57 transitions of 130 kev. The details of the calculations are given in Table I.

These two transitions seem to be identical with the 56.75- and 127.5-kev transitions, respectively, observed by Kyles and collaborators[1] in the decay of Ac^{228}. These authors also observed a 184.2 kev transition, identified as $M1$, on the basis of its K/L ratio, with an intensity ~1.2 times that of the 127.5-kev transition. It was assumed to be the cross-over transition from the second excited state of Th^{228}. Our results concerning the missing cross-over transition in the decay of U^{232} (<1 percent of 130-kev transitions) show that the 184.2-kev line is not the cross-over transition from the second excited state.

TABLE II. Alpha-branching ratios for U^{232}.

α energy (Mev)	Decay prob. (10^{-11} sec^{-1})	Theoretical branching ratio (without spin correction) R_T	Experimental branching ratio R_E	Experimental forbiddenness factor R_E/R_T	Theoretical spin correction factor
5.318[a]	8.3	0.680[a]	0.680	1	1 ($l=0$)
5.258	3.8	0.310	0.314	1	0.64 ($l=2$)
5.128	0.62	0.0505	0.0057	0.11	0.23 ($l=4$)
4.988	0.086	0.0070	0.00017	0.24	0.8 ($l=1$)

[a] See reference 9.

[9] F. Asaro and I. Perlman, Revs. Modern Phys. **26**, 456 (1954).

Recent unpublished studies by Asaro and Perlman[10] indicate a level of 330 kev in Th^{228}. By analogy with the decay schemes of neighboring even-even nuclei these authors concluded that this is a 1− state. We made a careful search for γ-ray transitions to the 2+ state and to the ground state from this level which revealed indeed the two corresponding peaks of 270 and 330 kev (Fig. 3).

From the coincidence spectrum shown in Fig. 4 we see that the 60-kev γ ray coincides with the 270-kev γ ray, but not with the 330-kev γ ray. We therefore conclude that a 330-kev state exists which probably has spin and parity 1−, although 2+ is also compatible with our experimental evidence. In the latter case a 140-kev transition to the 4+ state at 190 kev might be expected. A weak transition of this energy cannot be excluded on the basis of our experiments, since it would be masked by the 130-kev γ ray. The relative intensities of the 330- and 270-kev γ rays are $I_{\gamma 330}/I_{\gamma 270}=0.76$. The data used for the calculation of the absolute transition probabilities, assuming that both transitions are $E1$ transitions, are given in Table I. It follows that 57 percent of the transitions from this level go to the 2+ state and 43 percent to the ground state.[11]

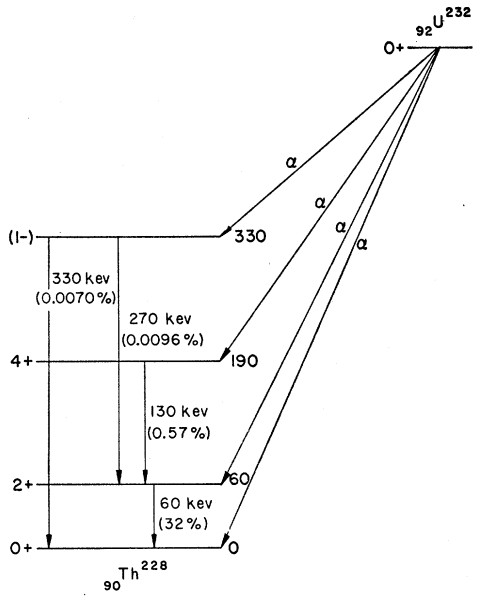

FIG. 5. Proposed disintegration scheme of U^{232}. The spin assignments of the 60-kev and 190-kev states are based on the measured internal conversion coefficients, whereas the spin assignment of the 330-kev state is tentatively made following Stephens, Asaro, and Perlman's systematics. (See reference 10.) However, an assignment of 2+ for this state is also compatible with the experimental evidence. From the experimentally determined relative intensities the number of transitions per alpha particle (in percent) were computed by using the value reported by Dunlavey and Seaborg (see references 5 and 9) for the 60-kev transition.

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

[11] It seems worthwhile mentioning here that H. B. Box and G. S. Klaiber [Phys. Rev. **95**, 1247 (1954)], who studied the

In Table II, α-particle branching ratios derived from theoretical transition probabilities[12] are compared with the experimental branching ratios calculated from the observed γ-ray intensities. In agreement with previous observations on even-even α emitters the α transition intensity to the 2+ state is approximately equal to that predicted by theory without taking the spin correction into account, whereas the transitions to the 4+ state and 1− state are reduced by factors 0.11 and 0.24 respectively, in contrast to the theoretical spin correction factors 0.23 and 0.84 respectively.[13]

The proposed decay scheme for U^{232} is shown in Fig. 5.

unconverted γ rays emitted from Ac^{228}, found lines of 278 and 336 kev, which they assigned to transitions from a 336-kev level to the first excited state and ground state of Th^{228}. However, their Fig. 3 indicates that in the Ac^{228} decay the 336-kev line is much stronger than the 278-kev line, in contrast to the intensity ratio found in the U^{232} spectrum. This may be due to the superposition of a 347-kev line corresponding to a transition from a 404 kev, postulated by the authors, to the 57-kev level.

[12] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952), p. 575.

[13] J. O. Rasmussen, Jr., University of California Radiation Laboratory Report UCRL-2431 (unpublished), and E. L. Church, private communication.

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Average Number of Neutrons per Spontaneous Fission of Cm^{244}†

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The average number of neutrons per fission, ν, for Cm^{244} has been measured by the manganous sulphate moderator-absorber system. The value 2.60 ± 0.11 has been determined.

A SAMPLE of Cm^{244} was purified by ion-exchange techniques and electroplated on a platinum foil of about 20 cm^2 area. The foil was rolled tightly and the heat output from it was measured calorimetrically. The calorimeter used was of the steady-state resistance bridge type used at the Mound Laboratory.[1] The accuracy of the present measurement was about one percent. A small portion of the original sample was pulse analyzed and the fraction of Cm^{242} was measured and the total heat output was corrected for the contribution from this source. From the alpha disintegration energy and half-lives for alpha emission and spontaneous fission,[2] the number of fissions in the sample was determined.

The number of neutrons emitted from the sample was determined by placing it at the center of a tank containing a saturated solution of manganous sulphate. After the Mn^{56}, which was produced by the capture of a neutron by Mn^{55}, was in equilibrium with the neutrons from the sample, the activity in the solution was determined with an immersion counter. The efficiency of the tank and counter system was measured by finding the ratio of counter events to neutrons from several calibrated polonium-beryllium sources. Since the tank was infinitely large to the polonium-beryllium neutron spectrum, no correction for spectral differences was necessary.

Corrections were made for the contribution to the number of neutrons from the spontaneous fission of Cm^{242}, using the value of $\nu=3.0\pm0.3$ determined by Barclay and Whitehouse.[3] This correction amounted to about 0.8 percent. An upper limit for the contribution from neutrons produced by the alpha particles on low-Z elements was determined by preparing a sample of Cm^{242} in the same manner and measuring an apparent ν in the same system. The value determined was 3.68, as compared with 3.0 measured by coincidence methods, and since the ratio of decay by alpha emission to decay by spontaneous fission is about twenty-five times larger for Cm^{242} than for Cm^{244}, and the energy of the alpha particles emitted by Cm^{244} is about 0.4 Mev lower than those of Cm^{242}, the error introduced from this source is estimated as about one percent. While the precision of the measurements is ±1 percent, the accuracy of the standards may be in error by as much as 3 percent.

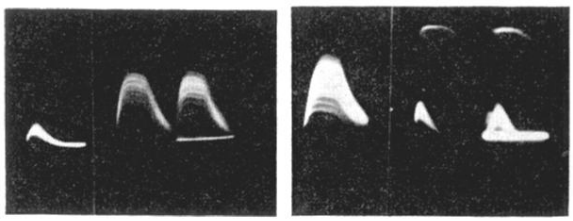
We are deeply indebted to S. G. Thompson for supplylng the source material and wish to express our thanks to J. Harper and L. Mann for their help with the electronic equipment.

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

[1] Eichelberger, Jordan, Orr, and Parks, Atomic Energy Commission Declassified Document AECD-3515 (unpublished) and other Mound Laboratory reports.

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

[3] F. R. Barclay and W. J. Whitehouse, Proc. Phys. Soc. (London) **A66**, 447 (1953).



(a) (b)

FIG. 1. $\alpha-L$ x-ray coincidences in U^{232}. (a) The first pulse reading from left to right is the L x-ray pulse from detector 1 with which the oscilloscope is triggered during the coincidence run. The second pulse is self-triggered and shows α-particle pulses in detector 2. The third pulse is the α-particle pulse in detector 2 shown while the oscilloscope was being triggered by the L x-ray pulse of detector 1. (b) The roles of the α and L x-ray pulses were reversed and again the first pulse shows the triggering pulse (α particle in detector 2), the second pulse shows the L x-rays in detector 1, self-triggered, and the third pulse shows coincidences between the triggering pulse and the signal (L x-ray in detector 1).

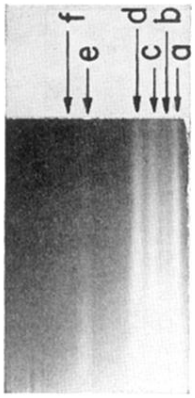


FIG. 4. Spectrum of photons coinciding with 60-kev γ rays of U^{232} as displayed with a gray wedge analyzer. This spectrum indicates that both the 330- and 270-kev γ transitions arise from a 330-kev level. Prominent lines are seen at $\sim$35 kev (a), $\sim$90 kev (K x-rays) (c), 130 kev (d), and 270 kev (e); f indicates the position of the 330-kev line which is missing in the coincidence spectrum. The position of the triggering 60-kev γ rays is given by b. The $\sim$35-kev line may be the 42-kev gamma ray of U^{233} which coincides with a 56-kev gamma ray.