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Alpha-Particle and Gamma-Ray Spectra of the U^{230} Decay Series*

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The radiations of the U^{230} family have been investigated with an electromagnetic alpha-particle spectrograph and gamma-ray scintillation counters. The following alpha groups were found: U^{230}—5.884 (67.2%), 5.813 (32.1%), 5.658 (0.7%); Th^{226}—6.330 (79%), 6.220 (19%), 6.095 (1.7%), 6.029 (0.6%); Ra^{222}—6.551; Em^{218}—7.127 Mev. The following gamma rays were seen: U^{230}—72 (0.75%), 158 (0.33%); 232 (0.24%); Th^{226}—112 (4.8%); 131 (0.4%), 197 (0.40%), 242 (1.2%); Em^{218}—609 kev (0.2%). The results are correlated into decay schemes, which are discussed with respect to current systematics and theory of complex alpha spectra and excited states of even-even nuclei.

INTRODUCTION

THE uranium isotope U^{230} is a 20.8-day α emitter which gives rise to a series of shorter-lived daughers.[1] The series is collateral to the uranium-radium family ($4n+2$ series) and joins the main family at Po^{214}(RaC′).[2]

$$\begin{tikzcd}
\text{Main family:} & U^{234} \arrow[r, "\alpha", "2.5\times10^{5}\text{ yr}"'] & Th^{230}(\text{Io}) \arrow[r, "\alpha", "8.0\times10^{4}\text{ yr}"'] & Ra^{226} \arrow[r, "\alpha", "1622\text{ yr}"'] & Em^{222}(\text{Rn}) \arrow[r, "\alpha", "3.83\text{ day}"'] & Po^{218}(\text{RaA}) \arrow[r, "\alpha", "3.05\text{ min}"'] & Pb^{214}(\text{RaB}) \arrow[d, "\beta^{-}\ 26.8\text{ min}"] & \\
 & & & & & & Bi^{214}(\text{RaC}) \arrow[d, "\beta^{-}\ 19.7\text{ min}"] & \\
 & & U^{230}\text{ family:} & U^{230} \arrow[r, "\alpha", "20.8\text{ day}"'] & Th^{226} \arrow[r, "\alpha", "30.9\text{ min}"'] & Ra^{222} \arrow[r, "\alpha", "38\text{ sec}"'] & Em^{218} \arrow[r, "\alpha", "0.019\text{ sec}"'] & Po^{214}(\text{RaC}').
\end{tikzcd}$$

This paper is concerned with the alpha and gamma spectra of this group of even-even alpha emitters. The early members of the series (U^{230}, Th^{226}) are sufficiently far removed from the closed-shell region at lead so that nuclear energy level spacings are small and hence excited states are appreciably populated by alpha decay.[3,4] As a result the spectra are complex and several low-energy γ transitions are observable. The lower members of the series show simpler spectra because energy level spacings are wider and the sharp dependence of alpha-decay lifetime upon energy effectively prevents the observation of transitions to excited states. Nevertheless evidence for the first excited states was obtained for both Em^{218} and Po^{214} (from the decay respectively of Ra^{222} and Em^{218}). Some of the data presented here have been summarized in a review article.[5]

EXPERIMENTAL

Preparation of sources.—The U^{230} was obtained from its Pa^{230} parent which had been made by the irradiation of thorium (Th^{232}) with protons in the 184-in. synchrocyclotron. Four preparations were made over a period of two years, the irradiation histories of which are summarized in Table I. Pa^{230}, which has a half-life of about 17 days, decays only to the extent of ~8% by β^{-} emission to U^{230} and ~92% by electron capture to long-lived Th^{230}.[6] At the proton energies employed for maximum thick-target yields of Pa^{230}, there are also formed in good yield 22-hr Pa^{228} and 36-hr Pa^{229}. These decay, respectively, to alpha-emitting thorium isotopes, 1.9-yr Th^{228} and 7340-yr Th^{229}. Because of their relatively short half-lives, Th^{228} and its daughters could interfere in the observation of the U^{230} series even after some chemical separation. This source of interference was minimized in the first three preparations by allowing the protactinium isotopes to decay for several days before they were isolated. The protactinium fraction then consisted predominantly of Pa^{230} and the long-lived Pa^{231}. The optimum waiting time for the growth

TABLE I. Irradiation data.

Irradiation number	Target	Proton energy	Length of irradiation
1	thorium	85 Mev	2.7 hr
2	thorium	145 Mev	1.6 hr
3	thorium	150 Mev	7.0 hr
4	thorium	100 Mev	4.0 hr 13.3 hr 10.0 hr

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

[1] M. H. Studier and E. K. Hyde, Phys. Rev. **74**, 591 (1948).

[2] All data presented without references were taken from Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).

[3] F. Asaro and I. Perlman, Phys. Rev. **87**, 393 (1952).

[4] S. Rosenblum and M. Valadares, Compt. rend. **235**, 711 (1952).

[5] I. Perlman and F. Asaro, Ann. Rev. Nuclear Sci. **4**, 157 (1954).

[6] M. H. Studier and R. J. Bruehlman, Argonne National Laboratory Report ANL-4252, February, 1949 (unpublished).

of U^{230} was about one month. In the fourth preparation the uranium was chemically isolated from the other irradiation products about one month after the irradiation.

The U^{230} was prepared as a source for the alpha-particle spectrograph by volatilizing from a tungsten filament onto a cold platinum plate. The tungsten filament was folded to make a V trough in order to "focus" the deposit into a narrow band.

For γ-ray measurements the uranium fractions were purified more rigorously than for α-particle analysis because one of the objectives was to search for γ rays of extremely low intensity. The uranium fraction was subjected to repeated ether extractions from ammonium nitrate solutions and to adsorption and elution from anion-exchange resin columns.

For many of the γ-ray analyses the various decay products of U^{230} were separated by collecting recoils resulting from the α-decay process. The techniques varied with the particular products to be collected and are described where the γ-ray analyses are described. The alpha spectrograph measurements are inherently slow and only the equilibrium mixture was dealt with.

Radiation measurement.—The electromagnetic spectrograph used for determining the α spectra has been described in other reports.[7,8] As in other studies photographic plates were used to record the alpha tracks, and alpha groups of known energy were used for energy standards.

The gamma-ray analysis employed a sodium iodide scintillation assembly, the output of which went into a 50-channel self-gated pulse-height analyzer. The crystal and photomultiplier tube were enclosed in a lead housing with a sample holder which allowed placement of the samples at five different distances from the crystal. The variable geometry was employed to help in the assignment of γ rays to particular members of the decay chain, as will be described.

Gamma-gamma coincidence measurements were made by using a single-channel analyzer to trigger the gate of the 50-channel analyzer. In this way the entire spectrum in coincidence with a selected photon energy could be recorded. Intensities were corrected for escape peaks[9] and crystal counting efficiencies.[10]

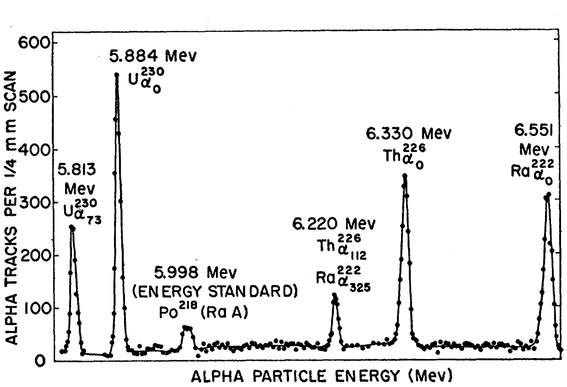

FIG. 1. Alpha spectrum of U^{230} family.

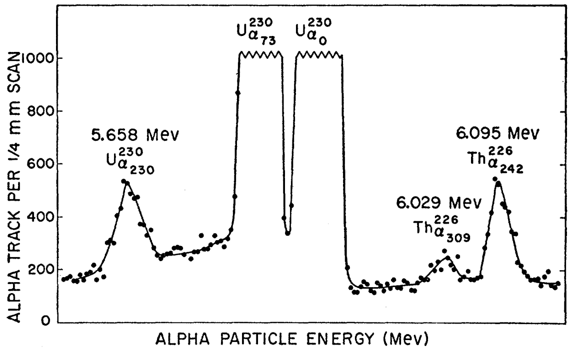

FIG. 2. Low-abundance alpha groups of U^{230} and Th^{226}.

RESULTS

Uranium-230

Alpha spectrum.—Since exposures on the alpha-particle spectrograph were long compared with the lifetimes of the U^{230} decay products all spectra taken included the equilibrium mixture. A part of the spectrum is shown in Fig. 1 and a longer exposure used to bring out low-intensity groups appears in Fig. 2.

The main alpha group (ground-state transition) of U^{230} was found to be at 5.884±0.005 Mev. The energy was measured on several runs by using either the main group of ThX[11] (5.681 Mev) or that of RaA (5.998 Mev)[11] as an energy standard. The previously reported value for the alpha-particle energy of U^{230} is 5.85 Mev determined with an ionization chamber.[12]

The alpha transition to the first excited state of Th^{226} is undoubtedly that at 5.813 Mev, designated U^{230} α_{73} in Fig. 1 to signify that this group leads to a state 73 kev above the ground state. The γ-transition energy is obtained from the difference of α-group energies plus the correction for the difference in recoil energy from the two alpha groups. From seven measurements of the alpha spectrum the difference between α_0 and α_{73} gave a best value for the γ transition of 72.6±0.5 kev.[13] Eight measurements of the abundance of α_{73} gave a value of (32.1±0.8)% of the total U^{230} α particles.

A third α group in low intensity ascribable to U^{230} was found at 5.658 Mev (see Fig. 2). The energy level of Th^{226} to which this group leads was found to be at

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

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

[9] P. Axel, Brookhaven National Laboratory Report BNL-271 T-44, September, 1953 (unpublished).

[10] M. Kalkstein and J. M. Hollander, University of California Report UCRL-2764, October, 1954 (unpublished).

[11] G. H. Briggs, Proc. Roy. Soc. (London) **A157**, 183 (1936).

[12] A. H. Jaffey, quoted by M. H. Studier and E. K. Hyde, Phys. Rev. **74**, 591 (1948).

[13] Recently Smith, Asaro, and Hollander [Phys. Rev. **104**, 99 (1956), following paper] measured *L*, *M*, and *N* conversion lines of this γ ray with a permanent-magnet spectrograph and obtained a more accurate energy (72.13±0.07 kev).

230±5 kev as a result of three measurements of the alpha spectrum. This agrees well with the presence of a 232-kev γ ray which will be mentioned further below. The intensity of α_{230} is $(0.7\pm0.15)\%$ of the total U^{230} alpha particles. There is good evidence that this α group is really an unresolved doublet, one group leading to a 4+ state and the other to a 1− state. The arguments for this conclusion appeared in part in an early publication concerning 1− levels in this region and will be reviewed briefly where the decay scheme is discussed. No other α groups ascribable to U^{230} could be found.

Gamma spectrum.—Three γ rays were found in the decay of U^{230}. The energies and abundances (shown in parentheses) are as follows: 72±2 kev $(0.75\pm0.11\%)$, 158±3 kev $(0.33\pm0.06\%)$, 232±3 kev $(0.24\pm0.05\%)$. The abundances are percentages of total alpha emission events.

The problem in determining the U^{230} γ spectrum is to distinguish its γ rays from those of Th^{226} and its decay products which grow into purified U^{230} with a 31-min half-life. In one experiment the γ-ray spectrum of the equilibrium mixture was measured and then that of Th^{226} and its products. The curves were normalized in terms of a peak at 325 kev (from Ra^{222}), and upon subtraction the U^{230} spectrum was obtained.

A more sensitive way of removing peaks due to Th^{226} and products is illustrated by Fig. 3. The uranium fraction was purified by ether extraction and the γ-ray spectrum measured soon after. Some of the peaks were due to U^{230} decay products because these grow in with a 31-min half-life and the relative intensities of some are greater than those of U^{230} γ rays. This spectrum is the broken-line curve of Fig. 3. After the daughters had grown in to a large extent the γ-ray spectrum was again measured (not shown in Fig. 3) and a subtraction of the second spectrum from the first gave the spectrum of the Th^{226} family. Then the 325-kev peaks in the first spectrum and that of the Th^{226} family were used to normalize the two curves. Subtraction of the normalized Th^{226} family spectrum gave the U^{230} spectrum (solid-line curve in Fig. 3). A small correction was made for the lag of the 325-kev γ ray which grew in with a 38-sec half-life.

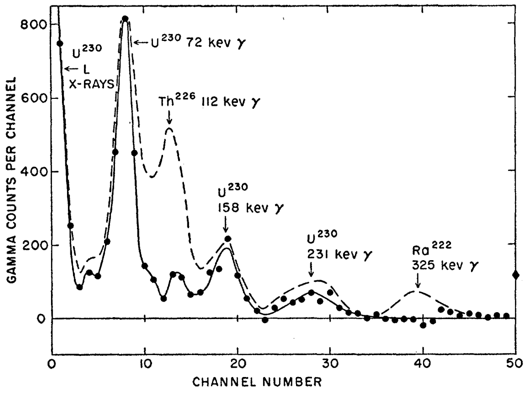

FIG. 3. U^{230} gamma-ray spectrum. - - - U^{230} family gamma spectrum measurement started 4.25 min after purification of uranium (6-min run). —— Net U^{230} gamma spectrum without daughter contributions.

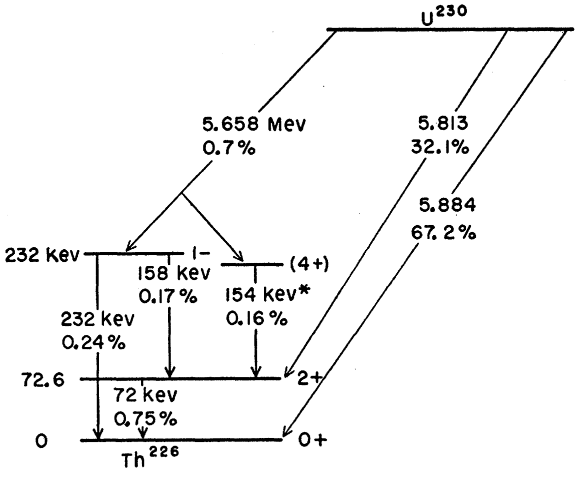

FIG. 4. U^{230} decay scheme. The transition marked with an asterisk is the probable assignment of a γ transition found by Smith, Asaro, and Hollander.[13]

This operation was repeated several times using ether extraction or anion columns to purify the uranium. As a result of the several measurements the γ-ray energies and intensities as already cited were obtained. Aside from the three photons definitely ascribed to U^{230} decay there appears to be a residual peak at ~110 kev when the Th^{226} peak at this energy is subtracted. Since this peak could receive contribution from scattered radiation from higher-energy γ rays or simply represent inaccuracy in the curve-subtraction process, it is better not to assume that this is a U^{230} γ ray.

Decay scheme.—The three alpha groups and three γ-ray lines agree well with the energy level scheme shown in Fig. 4. The interpretation of the levels, however, is not entirely straightforward.

The 73-kev level is undoubtedly the familiar 2+ first excited state expected in this region. The total conversion coefficient (value is 42) was found by comparing the total population of the 73-kev state with the measured γ-ray intensity, and this value corresponds with the expected $E2$ nature of the transition. Finally, in a study already reported,[14] alpha-gamma angular correlations for this transition corresponded with the requirements of a $0\xrightarrow{\alpha}2\xrightarrow{\gamma}0$ spin sequence.

A number of previous reports have dealt with the rotational spectra of even-even nuclei in the heavy-element region in which the second excited level of the rotational band has spin and parity 4+ and lies at an energy about 3.3 times that of the 2+ state.[15–17] In

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

[15] A. Bohr and B. R. Mottelson, Phys. Rev. **89**, 316 (1953); **90**, 717 (1953).

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

[17] A. Bohr and B. R. Mottelson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **27**, No. 16 (1953).

this case we would predict that the 4+ state would lie close to 230 kev which at first sight seems to be in excellent agreement with the observed energy level defined by the 5.658-Mev α group. However, for all 0+, 2+, 4+ sequences there has never been observed a crossover transition from the 4+ to the 0+ states and none would be expected. The 232-kev γ ray must therefore originate from a state other than 4+, and as shown in Fig. 4 the 232-kev level is assigned 1−. Confirmation of this assignment through alpha-gamma angular correlations has already been published.[15]

The question then remains whether there is any experimental evidence that a 4+ state also exists at ~230 kev as an unresolved doublet with the 1− state. The 232-kev γ ray can come only from the $1- \rightarrow 0+$ transition but, within the available resolution, a γ-ray of ~158 kev can arise from both $1- \rightarrow 2+$ and $4+ \rightarrow 2+$ transitions. It will be seen that the photon intensity of 0.33% is divided about equally between the $E1$ and $E2$ transitions (see Fig. 4). When the conversion coefficients are taken into consideration the alpha population of the 4+ state is calculated to be 0.43% and that of the 1− state (158 kev plus 232-kev transitions) is 0.44%. The sum of these, 0.87%, is to be compared with 0.7%, the measured unresolved α-group intensity populating both of these states. The agreement is considered satisfactory.

The first experimental evidence that there is more than one state at ~232 kev came from the comparison of γ rays of Th^{226} excited from two modes of decay. Grover and Seaborg[18] found in the β-decay of Ac^{226} the abundance ratio $\gamma_{159}/\gamma_{232}$ to be 0.85 and this experiment has been checked as part of the present study. However, the same relative γ-ray intensities from U^{230} α decay has the ratio 1.7. (These ratios, although measured in the same way, are used only for comparison and do not include corrections for counting efficiency.) Since it is unlikely that the β-decay process would populate both 1− and 4+ states we may conclude that only the 1− state is populated and therefore, from the α decay of U^{230}, half of the observed photon intensity at 158 kev arises from the 1− state and half from the 4+ state as indicated in Fig. 4.

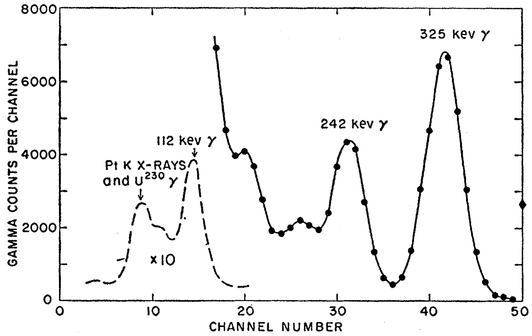

FIG. 5. Equilibrated U^{230} family gamma-ray spectrum.

A selective view of the $4+ \rightarrow 2+$ transition was obtained by Smith, Asaro, and Hollander[13] in this laboratory in measuring the conversion electron spectrum of the U^{230} series with a permanent magnet spectrograph. Among the lines seen were L_{II} and L_{III} lines of a γ ray of 154.3±0.3 kev, while the L_{I} line was missing. The absence of the L_{I} line strongly indicates $E2$ character and therefore that this is the $4+ \rightarrow 2+$ transition. Since the observed lines were weak one would not expect to have seen the $1- \rightarrow 2+$ $E1$ transition because the L-shell conversion coefficient is some fiftyfold lower. It would appear from these measurements and from our value, 158±3 kev, for the γ-ray energy, that the 4+ state lies a few kev below the 1− state.

One further point concerning the doublet at 230 kev is the alpha-gamma angular correlation measurements previously reported.[15] The 70-kev γ ray showed a well-defined $0\xrightarrow{\alpha}2\xrightarrow{\gamma}0$ correlation and the 230-kev γ ray followed clearly the $0\xrightarrow{\alpha}1\rightarrow 0$ form. However, the alpha-gamma angular dependence of the 160-kev γ ray had little if any anisotropy. This is consistent for a mixture of radiations from the sequences $0\xrightarrow{\alpha}4\xrightarrow{\gamma}2$ and $0\xrightarrow{\alpha}1\xrightarrow{\gamma}2$.

Radium-222

Gamma rays.—A single γ ray of 325±3 kev has been found from the α decay of Ra^{222} and is attributed to the de-excitation of the first excited state of Em^{218}. Figure 5 shows part of the γ-ray spectrum of the U^{230} family among which is a fairly prominent peak at 325 kev. The energy for the γ ray is the weighted average of four measurements. It has been assigned to Ra^{222} decay from the following experiments.

A thin sample of Th^{226} was prepared by collecting under vacuum the recoiling atoms from U^{230} decay. The Th^{226} decay product, Ra^{222} (38 sec half-life), could be collected in an interesting manner which permitted immediate γ-ray analysis: The scintillation crystal and photomultiplier tube are enclosed in the standard fashion, in an aluminum can, which, if not grounded, floats at a potential of over 500 volts negative when the photomultiplier is on. This potential effectively collects recoils from an alpha emitter placed below the crystal. The Th^{226} sample was alternately placed below the crystal and removed at one-minute intervals. During the time the Th^{226} was out of the counting chamber a switch was tripped to allow the γ spectrum to register. The operation was repeated to record a total of 20 minutes' counting time. The background count was taken for 20 minutes and subtraction gave the curve shown in Fig. 6. It is seen that the only definite γ ray present is that at 325 kev; in particular, the prominent γ rays at 112 kev and 242 kev have disappeared and it will be seen that these are due to Th^{226}.

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[18] J. R. Grover and G. T. Seaborg (unpublished data, 1954).

A further check that the 325-kev γ ray belongs to Ra^{222} (or its daughters) was obtained by determining the half-life of the 325-kev peak after collecting recoils from Th^{226} in the manner already described. A value of 36±2 sec was obtained which agrees well with the 38-sec half-life reported by Studier and Hyde.[1] The α-activity of a recoil sample from Th^{226} decay collected in vacuum gave a half-life of 37.5±2 sec.

That the 325-kev γ ray belongs to the decay of Ra^{222} and not to its daughter was determined in the following manner. It was already mentioned that the metal can in front of the crystal serves as a collector of recoiling nuclei from alpha-emitting samples placed near it. If a sample containing a family of alpha emitters is placed at different distances from the crystal then a particular γ ray will show a different intensity pattern depending upon whether it comes from the primary α emitter or one of the daughters. If the γ ray arises from the parent substance the measured intensity should vary in first approximation simply as the solid angle subtended between sample and the crystal. If, however, the γ ray arises from one of the α-decay products, the recoil collection by the crystal housing will tend to make the intensity independent of the sample position for the component of the recoils which leave the plane of the sample in the direction of the crystal.

TABLE II. Assignment of 325-kev γ ray.

Sample position	Calibrated solid angle (Relative)	Calc relative intensities from recoils	Relative intensities of 325-kev γ ray from Th^{226} sample	Relative intensities of 325-kev γ ray from Ra^{222} sample
1	1	1	1	1
2	0.22	0.56	0.56	0.28
3	0.10	0.49	0.45	0.10

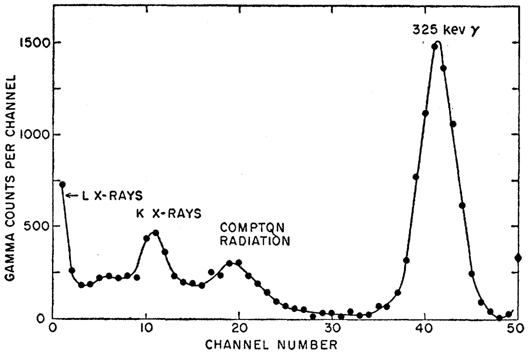

FIG. 6. Ra^{222} family gamma-ray spectrum.

The relative solid angle for three different sample positions was determined using the 60-kev γ ray of Am^{241} as a monitor, and these are shown in Column 2 of Table II. Column 3 shows the calculated[19] relative intensities of a γ ray which arises from a recoiling product. The fourth column shows the relative intensity of the 325-kev peak measured in a sample of Th^{226} which had been prepared by collection of recoils in vacuum from a U^{230} sample. Clearly the 325-kev γ ray follows the intensity pattern for a product of Th^{226} and since this fact was already known the experiment checks the validity of the method. Next samples of Ra^{222} were collected from Th^{226} under vacuum and quickly placed in the scintillation spectrometer. In a series of carefully controlled experiments in which the three sample positions were included and the counting rates of the 325-kev peaks corrected for decay, the results as shown in the last column of Table II were obtained. Since the relative intensities follow the solid angle dependence the γ ray must come from Ra^{222} decay and not from a decay product.

Still another check was made later[15] on the assignment of the 325-kev γ ray to Ra^{222}. A thin scintillation crystal was used as a rough alpha energy measuring device and was coupled to a single-channel pulse-height discriminator which in turn served as a gate for the multichannel γ-ray analyzer. The coincidence experiments showed that the 325-kev γ ray was in coincidence with alpha particles of energy about 0.33 Mev lower than the known energy of the ground-state transition of Ra^{222}. This α-energy setting is nowhere near the energy of Em^{218} or Po^{214} α-particles and therefore the γ ray must come from Ra^{222} decay, since it is already known that it cannot arise from earlier members of the decay chain.

K x rays were observed in the Ra^{222} gamma spectrum and these presumably arose from the conversion of the 325-kev gamma ray. The abundance of these *K* x-rays corresponds to a *K*-conversion coefficient of 0.08, indicating an *E*2 transition. Later alpha-gamma angular correlation measurements[15] confirmed this assignment. The 325-kev γ ray of Ra^{222} was found in 3.6 percent abundance. The sum of gamma ray, *K* conversion, and the theoretical *L* conversion yields a population of 4.4±1 percent to the 325-kev state. It will be seen that the particular alpha group leading to this state could not be resolved from another (see Fig. 1), so that the γ-ray measurement is the only means at present for obtaining the intensity of the alpha group. All other gamma-ray abundances described in this paper were

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[19] Recoils from U^{230} alpha decay were collected in vacuum on a plate masked so that the recoils did not enter at angles more acute than 8°. If we assume the ranges of recoils following U^{230} and Th^{226} alpha decay are equal, the fraction of Ra^{222} activity which escapes the collecting plate is

$$\int_{8^\circ}^{90^\circ}\left(\frac{1-\sin\theta}{2}\right)d\theta \Big/ \int_{8^\circ}^{90^\circ} d\theta = 0.15.$$

The angle θ is the angle of incidence of the recoil fragments in entering the collecting plate. Assuming that all of the escaping recoils are attracted to the metal container which houses the crystal and that the crystal subtends a solid angle of 45% of 4π sterad for any gamma ray emitted by atoms on the housing, we find, for the three sample positions indicated in Table II (solid angle for position $1 = 10.8\%$ of 4π sterad), that the expected relative counting rates for gamma rays following Ra^{222} decay are 1:0.56:0.49.

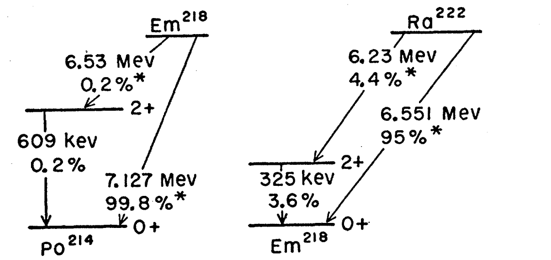

FIG. 7. Decay schemes of Em^{218} and Ra^{222}. The abundances marked with an asterisk were deduced from the gamma-ray abundances.

measured relative to the 325-kev gamma abundance, and the listed limits of error are also relative to the intensity of this gamma ray. This value is accurate to about 20%.

Alpha spectrum and decay scheme.—The alpha peaks of Ra^{222} are shown in Fig. 1 and the energy for Ra^{222} α_0 is 6.555±0.010 Mev. The previous value reported for this alpha emitter is 6.51 Mev, which was obtained with an ionization chamber.[12] The alpha group leading to the 325-kev state falls at nearly the same position as a prominent α group of Th^{226} and the two cannot be resolved. From the γ-ray intensity measurements it can be shown that $\frac{1}{5}$ of the peak at 6.220 Mev is due to Ra^{222} α_{325} and the other $\frac{4}{5}$ belongs to Th^{226} α_{112}. The decay scheme summarizing the information on Ra^{222} decay is shown in Fig. 7. Other excited states of Em^{218} could not be seen in the experiments presumably because they lie at a high level and hence would be populated only to a very slight extent.

Thorium-226

Gamma rays.—The Th^{226} sample was made by collecting recoils in vacuum from a U^{230} preparation. The plate was covered with tape to prevent loss of Th^{226} products and the spectrum of the entire family recorded. The contribution by Ra^{222} (and its products) was subtracted after normalizing the Ra^{222} spectrum to that of the Th^{226} family in terms of the 325-kev γ ray. The resulting spectrum of Th^{226} alone is shown in Fig. 8.

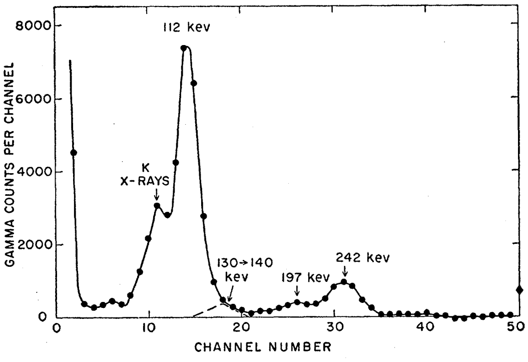

FIG. 8. Th^{226} gamma-ray spectrum.

Of the four γ rays indicated, that at 130 kev had to be resolved with the aid of the gamma-gamma coincidence spectrum gated by the pulses from the 112-kev γ ray. This curve shown in Fig. 9 proves that γ rays of ~130 kev and ~190 kev are in coincidence with the 112-kev γ ray but that the 242-kev γ ray is not.

From a number of experiments the best energy and abundance values for Th^{226} γ-rays are: 112 (±3) kev, 4.8 (±0.4)%; 131 (±5) kev, 0.4 (±0.1)%; 197 (±10) kev, 0.40 (±0.05)%; 242 (±3) kev, 1.2 (±0.1)%. As expected from the 112-kev $E2$ transition, L x-rays were found in high abundance.

Alpha spectrum and decay scheme.—The alpha groups of Th^{226} are seen in Figs. 1 and 2. The main group (ground-state transition) is at 6.330 (±0.010) Mev which compares with the ionization chamber measurement,[12] 6.30 Mev. The group at 6.220 Mev combines α_{112} of Th^{226} and α_{325} of Ra^{222} as already discussed. If correction is made for the Ra^{222} contribution to this peak the intensity of Th^{226} α_{112} is 19 (±1.5%). The energy difference between this state populated by α_{112} and the ground state turns out to be 112±3 kev from the alpha spectrum, just as was found for the γ ray. As indicated in the decay scheme (Fig. 10) the 112-kev level is the familiar 2+ first excited state. The conversion coefficients of the 112-kev γ ray agree with its expected $E2$ character. The γ-ray intensity is 4.8%, the population of the state is 19%, and therefore the total conversion coefficient is 3.0 which upon comparison with theoretical conversion coefficients rules out an $E1$ assignment. From the total K x-ray intensity it was found that the *maximum* contribution to the conversion coefficient from K conversion is 0.4. This information serves to rule out $M1$ character; the L-shell conversion coefficient does not distinguish well between $E2$ and $M1$ at this energy. The $E2$ assignment was later confirmed by angular correlation measurements.[15]

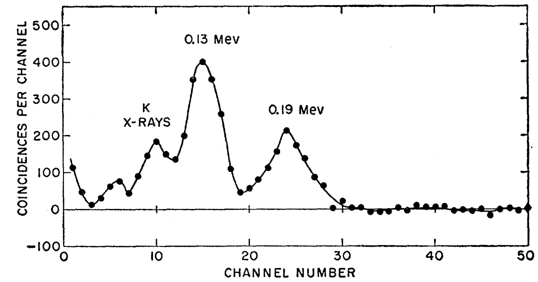

FIG. 9. Th^{226} family gamma-ray spectrum in coincidence with 112-kev gamma ray.

An α-group at 6.095 Mev was assigned to Th^{226} since its energy separations from Th^{226} α_0 and α_{112} agree well with the 242- and 130-kev γ rays of Th^{226}. In addition the 130-kev γ ray was shown to be in coincidence with the 112-kev γ ray. Since the 242-kev state populates by γ emission both the 0+ and 2+ states it can have spin 1 or 2 only, and from conservation laws in the α-decay process the assignment more specifically is 1− or 2+. Since this level bears the same relationship to other levels as the 1− state in Th^{228} decay it was assumed to have that assignment here. The assignment was later proved by alpha-gamma angular correlation measurements.[15] The best abundance for Th^{226} α_{242} based on four measurements is 1.7 (±0.15)% of the total Th^{226} α particles.

The α group at 6.029 Mev was assigned to Th^{226} because the energy separation from Th^{226} α_{112} agrees very well with the 197-kev γ ray and this γ transition is in coincidence with the 112-kev γ ray. The best energy for the level populated by this group is 309 kev, the abundance of the α group is 0.58 (±0.06)%, and the abundance of the 197-kev γ ray is 0.40% of the total Th^{226} α-decay rate. The conversion coefficient is therefore 0.4, a value which agrees with an $E2$ assignment and not with $M1$ or higher order electric multipole radiations. The evidence agrees well with the expectations for the second excited member of the rotational band and hence is designated 4+ in Fig. 10.

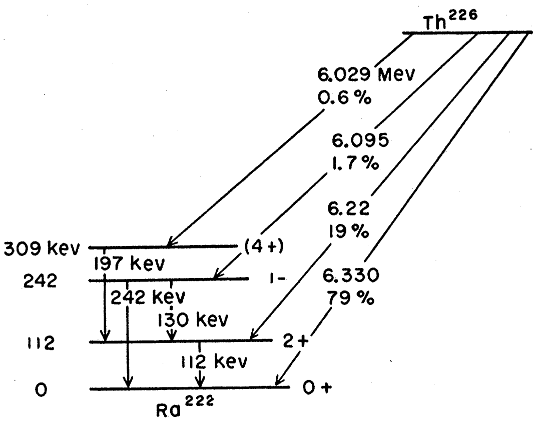

FIG. 10. Th^{226} decay scheme.

Emanation-218

The α group at 7.127 (±0.010) Mev is assigned to Em^{218}; the previously reported energy is 7.12 Mev.[12] The high-energy part of the γ-ray spectrum of Th^{226} and products recorded in Fig. 11 shows the 325-kev γ ray of Ra^{222} and another peak at 609±6 kev found in (0.20±0.05)% of the Em^{218} α-decay events. The assignment to Em^{218} decay is based on the following evidence: The γ ray belongs to a member of the series below Th^{226} because it was found in the Ra^{222} recoils collected from Th^{226} decay. When a thin KI crystal was used as an α-particle spectrometer[15] and a selected α-energy band at ∼0.61 Mev below the main Em^{218} group (7.13 Mev) was used in a coincidence measurement, it was found to be in coincidence with the 609-kev γ ray. Presumably the 609-kev state of Po^{214} seen here from the α decay of Em^{218} is that which leads to the well-known γ ray of this energy for the β-decay of Bi^{214}. The alpha group of Em^{218} leading to this state could not be seen because of its low intensity and close proximity to the main group of Ra^{222}.

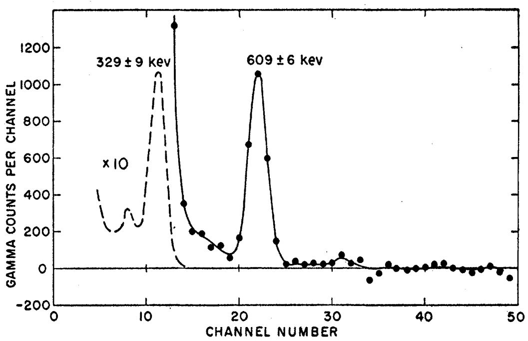

FIG. 11. Th^{226} family high-energy gamma-ray spectrum through 4.03-g/cm² Pb absorber.

DISCUSSION

A graphical summary of energies and intensities of the α groups which have appeared in the U^{230} series is given in Fig. 12. The ordinate scale showing intensities was made logarithmic in order to accommodate the large range.

The region starting with U^{232} and proceeding to lower elements marks the departure from the extreme uni-

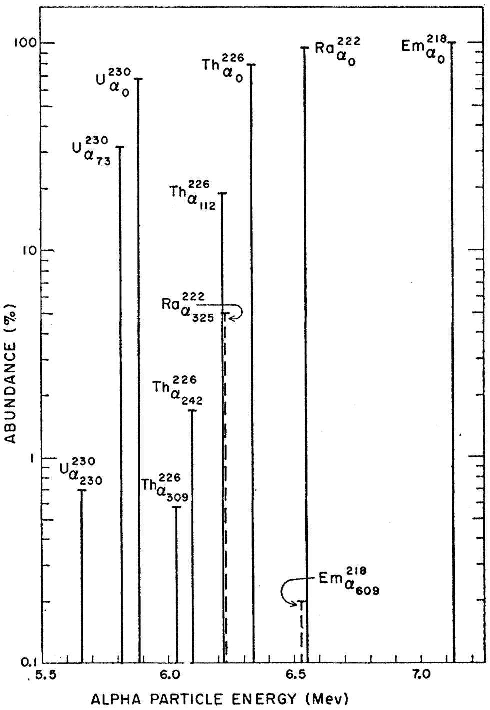

FIG. 12. U^{230} family alpha-particle energies and abundances.

TABLE III. γ-ray lifetimes of the U^{230} family.

Excited states of	Transition	Energy (kev)	Half-life (millimicro-seconds)
Th^{226}	$2+\rightarrow 0+$	73	<1.5
Ra^{222} (Th^{226})	$1-\rightarrow 0+$	230–240	<1.2
Ra^{222}	$(4+)\rightarrow 2+$	200	<1.5
Th^{226}	$1-\rightarrow 2+$ $(4+)\rightarrow 2+$	160	<1.5
Ra^{222}	$2+\rightarrow 0+$	110	<1.4
Em^{218}	$(2+)\rightarrow 0+$	325	<0.8

formity noted for the α spectra of the heavier even-even α emitters. For the heaviest elements the Bohr-Mottelson rotational spectra are nearly "pure" in the sense that the energy level spacings for the even states $0+$, $2+$, $4+\cdots$ follow closely the $I(I+1)$ dependence, and indeed the absolute values of the energies are almost identical. The energy for the first excited state ($2+$) in this region is somewhat over 40 kev. Examination of the decay schemes of the U^{230} family show progressive increases in energy of the first excited states: Th^{226}, 73 kev; Ra^{222}, 112 kev; Em^{218}, 325 kev; Po^{214}, 609 kev.

Bohr[15] has discussed the purity of rotational spectra and predicted that such spectra would be perturbed as the rotational motion becomes sufficiently rapid so that the particle structure of the nucleus can no longer follow adiabatically. According to Bohr[15] the first correction term is negative and follows an $I^2(I+1)^2$ dependence, which means that the higher rotational states become proportionally lower. Smith and Hollander[20] have obtained accurate energies for the first two excited states of Pu^{238} (from Cm^{242} decay) and from these evaluated the coefficients A and B for the rotational spectrum expression

$$E_{\mathrm{rot}}=A[I(I+1)]-B[I^2(I+1)^2].$$

The purity of the rotational states is evidenced by the finding that the coefficient B has the small value 0.0035 kev as compared with $A=7.37$ kev. The "$I^2(I+1)^2$" correction amounts to only 1% for the $4+$ state but would be 2.0% and 3.4% for $6+$ and $8+$ states. When applied to the $6+$ state[21] of Pu^{238} and the recently found[21] $8+$ state, the agreement between experiment and calculation was exact within the limits of error of the data. In the present study, the highest spin states observed were the $4+$ states in Th^{226} and Ra^{222} so that the coefficients A and B can only be calculated without any means of checking the adequacy of the simple "$I^2(I+1)^2$" correction. The observation of $6+$ states would be of great value in this region because the indicated correction is large and therefore the point at which this term is no longer adequate could be determined sensitively.[22] For the $4+$ state of Th^{226} the indicated correction is already 6% and for Ra^{222}, 23%. On this basis the $6+$ state of Th^{226} should lie at about 435 kev and would be seen in terms of the $6+\rightarrow 4+$ γ ray of $\sim$205 kev.

Another point of interest in the U^{230} family is the appearance of $1-$ states in two of the members. The occurrence of an odd parity state in an even-even nucleus at an excitation of only a few hundred kev has not yet received a verifiable explanation. A suggestion has been made, however,[23] that this state may have the same intrinsic structure as the ground state but represents a collective distortion in which the nucleus is pear-shaped. On this basis this state would belong to the $K=0$ configuration of the ground state, a supposition which can be checked by comparing the γ-ray intensities leading to the $0+$ and $2+$ states. Following the treatment of Alaga, Alder, Bohr, and Mottelson,[24] the reduced transition probabilities of γ rays leading from a particular state to two members of a rotational band would depend simply on the geometrical factors of the transition and can be expressed in terms of the vector addition coefficients. In the present case the two γ rays are electric dipole transitions ($1-\rightarrow 0+$, 0[25] and $1-\rightarrow 2+$, 0) and the question is whether the $1-$ state belongs to the $K=0$ structure or $K=1$. If the $1-$ state has $K=0$ the reduced transition probability ratio of $1-\xrightarrow{\gamma}0+/1-\rightarrow 2+$ should be 0.5; if $K=1$ for the $1-$ state, the ratio should be 2.0. It turns out that the experimental values for the corresponding states in Th^{226} and Ra^{222} are respectively 0.43 ± 0.08[26] and 0.48 ± 0.15. Clearly from this evidence the $1-$ state has $K=0$ and presumably has the same particle structure as the familiar $0+$, $2+$, $\cdots$ group of states. Other cases in which $1-$ states have been observed show the same behavior and have been dealt with in a separate publication.[27]

The even states of rotational bands such as dealt with in the present study are joined by cascading $E2$ transitions which are expected to be about 100-fold

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[20] W. G. Smith and J. M. Hollander, Phys. Rev. **101**, 746 (1956).

[21] Asaro, Harvey, and Perlman (unpublished data, 1955).

[22] The observations [Stephens, Asaro, and Perlman (to be published)] of $6+$ states following the alpha decay of Th^{230} and perhaps Th^{228} indicate that the simple "$I^2(I+1)^2$ correction" leads to values of the $6+\rightarrow 4+$ transitions which are 13% and 23% too low, respectively.

[23] A. Bohr (private communication of a suggestion by R. Christy).

[24] Alaga, Alder, Bohr, and Mottelson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **29**, No. 9 (1955).

[25] This notation expresses the spin and parity on the left side of the comma and the quantum number on the right side.

[26] More recent work [Stephens, Asaro, and Perlman (unpublished data, 1956)] on the decay scheme of Ac^{226} has shown that the ratio of the reduced transition probabilities for Th^{226} is 0.50 ± 0.05.

[27] Stephens, Asaro, and Perlman, Phys. Rev. **100**, 1543 (1955).

faster than $E2$ transitions governed by single-particle states.[17] There is abundant evidence that such transitions are indeed fast,[28–31] and the following table gives some upper limits to transitions of this type among members of the U^{230} family. Also included are some $E1$ transitions. The alpha-gamma fast-coincidence measurements were made by Strominger and Rasmussen.[32]

ACKNOWLEDGMENTS

We wish to acknowledge the participation of Dr. Louis Slater in the early parts of this study, and helpful discussions with Dr. Frank S. Stephens, Jr.

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[28] Experimental results are summarized in reference 18.

[29] D. Engelkemeir and L. B. Magnusson, Phys. Rev. **94**, 1395 (1954).

[30] A. W. Sunyar, Phys. Rev. **98**, 653 (1955).

[31] E. L. Church and A. W. Sunyar, Phys. Rev. **98**, 1186 (1955).

[32] D. Strominger and J. O. Rasmussen, University of California Radiation Laboratory Report UCRL-3157, June, July, and August, 1955 (unpublished).

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Electron Spectrum of the U^{230} Decay Series*

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The energies of the first excited states of Th^{226}, Ra^{222}, and Em^{218} have been measured as 72.13 ± 0.06, 111.1 ± 0.3, and 324.6 kev with photographic-recording beta-ray spectrographs. The energy of the second excited state in Th^{226} has been measured as 226.4 kev, and a comparison is made with predictions of the Bohr-Mottelson nuclear model.

THIS note will describe the results of a study of the conversion-electron spectrum of the U^{230} series, in which precision measurements have been made of the energies of the first excited states of Th^{226}, Ra^{222}, and Em^{218}, and a value also reported for the second excited state of Th^{226}. This work is part of a program concerned with a study of the detailed behavior of low-lying rotational states in heavy nuclei.[1]

The alpha- and gamma-ray spectra of the U^{230} family have been studied by Asaro and Perlman,[2] who reported from alpha-particle spectrograph and scintillation spectrometer measurements the energies of the first excited states in Th^{226}, Ra^{222}, and Em^{218} as 72.6 ± 0.5, 112 ± 3, and 325 ± 3 kev, respectively. They also find that the $4+$ level in Th^{226} lies near the $1-$ level at 232 ± 3 kev. Grover and Seaborg[3] also found the $1-$ level at 232 kev from Ac^{226} beta decay.

The present measurements have been made with two 180° photographic recording permanent-magnet spectrographs of effective field strengths 52 and 98 gauss; the instruments and their calibration have been described previously.[1]

The U^{230} was obtained by separation from its parent, Pa^{230}. The Pa^{230} had been prepared by irradiation of a piece of thorium metal with 100-Mev protons in the 184-in. cyclotron. U^{232} was also formed in this bombardment following beta decay of Pa^{232}. About one month after the irradiation the thorium target was dissolved, and a uranium fraction was separated by selective stripping of the activities from a Dowex A−1 resin column with hydrochloric acid solutions. Further purifications of the uranium fraction were made by ether extractions from $0.1N$ HNO_3 solutions saturated with ammonium nitrate.

The pure uranium fraction was evaporated to dryness then taken up in 500 μl of $0.4M\,(NH_4)_2C_2O_4$ in a special plating cell[1] from which the uranium was electrodeposited upon a 10-mil platinum wire. The active wire was then mounted in the appropriate spectrograph camera.

Although the spectrographs have been extensively calibrated, it was decided to use an internal standard in these experiments to gain the maximum precision; for this purpose, the 120-day beta emitter $_{69}Tm^{170}$ was used. The energy of the gamma ray of Tm^{170} has recently been measured by Hatch and DuMond[4] (using a 2-meter diffraction spectrometer) to be 84.26 ± 0.02 kev. This activity makes a very good standard both because its conversion electrons are quite similar in energy to those of U^{230} and also because thulium is easily electroplated.

Two independent measurements were made in the 52-gauss spectrograph of the energy of the U^{230} gamma ray. In the first, the Tm^{170} was initially plated upon

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[1] W. G. Smith and J. M. Hollander, Phys. Rev. **101**, 746 (1956).

[2] Frank Asaro and I. Perlman, Phys. Rev. **104**, 91 (1956), preceding paper.

[3] J. R. Grover and G. T. Seaborg (unpublished results, 1954).

[4] E. Hatch and J. W. M. DuMond (private communication, September, 1955).