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DECAY SCHEMES AND NUCLEAR SPECTROSCOPIC STATES IN THE HEAVY-ELEMENT REGION

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DECAY SCHEMES AND NUCLEAR SPECTROSCOPIC STATES
IN THE HEAVY-ELEMENT REGION

Frank Samuel Stephens, Jr.
(Thesis)

June 1955

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Decay Schemes and Nuclear Spectroscopic
States in the Heavy-Element Region

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June 1955

ABSTRACT

Using principally a coincidence circuit coupled to a 50-channel pulse-height analyzer, a study of the radiations from the following isotopes was made: 100^{254} , $99^{253, 254}$, $Cf^{246, 249, 250, 252}$, $Am^{239, 243}$, $Np^{237, 238}$, $U^{230, 235}$, $Th^{226, 228, 229, 230, 231}$, Ac^{225} , $Ra^{222, 224, 225, 226}$, Fr^{221} , Em^{218} , At^{217} , Po^{213} , Bi^{213} , Tl^{209} , and Eu^{154} . In most cases decay schemes or at least partial decay schemes were suggested.

The data on the even-even isotopes were found to fit in well with the previously existing systematics except for a group of low-lying 1^- states in the radium-thorium region. The information available on the five members of this group has been discussed in some detail. Regularities have been noted in the energy levels of odd mass nuclei in this region, and a preliminary systematics has been suggested.

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Decay Schemes and Nuclear Spectroscopic States in the Heavy-Element Region

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I. INTRODUCTION

A study of the decay schemes of a considerable number of nuclei in the heavy-element region has been undertaken in the hope that a more detailed knowledge of the energy levels involved will lead to a better understanding of their nature and therefore of the nucleus itself. In the even-even nuclei, where energy level regularities are well established,¹⁻⁵ an attempt was made to extend these regularities both by extending the region over which information is available and by looking in greater detail at several members within this region. In the odd mass number case decay schemes were examined with a view toward finding some kind of order among the many energy levels known to exist. Although not intentionally, no decays were studied which led to levels in odd-odd nuclei.

II. EXPERIMENTAL TECHNIQUES

The primary instrument used in this investigation was a coincidence circuit coupled to a 50-channel pulse-height analyzer. Scintillation detectors for all types of radiation from nuclei were either available or developed, making possible a wide variety of

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experiments. In conjunction with this unit instruments of high resolution, in particular an electromagnetic alpha particle spectrograph, were used wherever possible.

A. Scintillation Spectroscopy

Scintillation spectroscopy has recently become quite useful as a method of detecting radiations due to its short resolving time, high efficiencies, and improved energy resolution. Briefly it consists of a phosphor, which fluoresces when struck by an ionizing particle, in optical contact with a photomultiplier tube. The fluorescence radiation is converted in the photomultiplier tube to an electronic pulse which may subsequently be amplified and counted. If the light collection is uniform and the amplification linear, the size of the pulse will be proportional to the energy dissipated in the phosphor. Thus if the output pulses are pulse-height analyzed the energy spectrum of the incident particles is obtained. The resolving time or length of the output pulse is dependent on the duration of the fluorescence in the phosphor, which for those commonly in use varies from roughly 10^{-8} to 10^{-6} second. By selection of a scintillator which transmits its own fluorescence radiation, thicknesses of several inches may be used, permitting counting efficiencies of 100 per cent even for gamma rays of up to a few hundred kev.

1. Gamma ray detector. --Thallium activated sodium iodide is almost universally used for the detection of gamma rays. Since it

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is quite hygroscopic and must therefore be protected from the atmosphere, a special mounting technique developed by Borkowski was used. This consists of sealing the crystal in an aluminum can, with a glass window over one face to permit optical contact with the photomultiplier tube. Magnesium oxide is placed around the rest of the crystal to act as a light reflector. Single crystals of sodium iodide, 1.5 inches in diameter by 1 inch thick were used. These were at first mounted by us but later commercially packaged crystals became available which were quite satisfactory. One crystal was obtained which had a beryllium window instead of the usual aluminum one in order to permit the counting of lower energy photons (10 to 20 kev) which would be absorbed by the aluminum. RCA-5819 and Dumont-6292 photomultiplier tubes were used, of which the latter seemed somewhat superior on the basis of improved resolution and lower noise levels. Optical contact between the glass window of the crystal and the phototube surface was achieved with white vaseline. The optimum energy resolution (which is defined as the full width of a peak at half maximum divided by the energy) of crystals mounted in this manner was around 7 percent for the 0.66-Mev peak of Cs¹³⁷.

When a gamma scintillation spectrometer of this type is used for quantitative measurements, corrections must be made for escape peak losses and for counting efficiency. The escape peak is about 30 kev lower in energy than the main photopeak and is caused by the escape of an iodine K x-ray from the surface of the crystal. Calculations of the magnitude of this effect as a function of geometry and energy have been made by Axel,⁶ and these seem to be in good

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agreement with the experimentally observed values. Counting efficiency as used here will mean the ratio of events in the photopeak and escape peak to total events in the solid angle subtended by the crystal. Several processes tend to reduce this number. The most important of these are Compton scattering where the scattered photon escapes from the crystal and passage of the initial gamma ray through the crystal with no interaction at all. Kalkstein and Hollander⁷ have reviewed this subject in a recent publication and curves are presented, again as a function of energy and geometry, which seem to be in good agreement with all the experimental data. Unless specifically noted otherwise, all gamma ray intensities have been corrected in accordance with the references cited here.

2. Alpha particle detector. --Several scintillators were used for detecting alpha particles, depending on the geometry and resolution desired. For high geometry with essentially no energy resolution zinc sulfide screens sprayed directly onto RCA-5819 photomultiplier tubes were used quite successfully. Geometries of 35 to 40 percent could be obtained, with no noise or background interference from gamma or beta radiation. In several cases, however, sufficient energy resolution was needed to separate successive members of a decay family. For this purpose very thin sodium iodide or potassium iodide (thallium activated) crystals prepared in the following manner were used. The crystals were about 5 to 10 mm square by 1 mm thick, and were cleaved on all six sides. It is particularly important for

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good resolution that the surface facing the alpha particles be a near-perfect crystalline face. The crystals were mounted in myvawax onto a lucite light pipe about 1/8 inch thick by 1 to 1.5 inches in diameter, which was then fastened with a vaseline seal to a Dumont-6292 photomultiplier tube. It was found necessary to employ a collimator such that alpha particles did not strike near the edge of the crystal surface. When sodium iodide was used a vacuum system was employed, with a liquid nitrogen trap between the pump and the chamber, and silica gel within the chamber to absorb moisture, but even so the energy resolution dropped off severely over a period of a few days to a week. It was for this reason potassium iodide was later used. Mounted in the same manner and operated in air, this type of crystal showed little deterioration over a period of about a month. The optimum expected energy resolution of sodium iodide can be obtained by using the 7 percent obtained for a 0.66-Mev photon, and the fact that resolution should vary as the inverse square root of energy. This gives 2.5 percent for a 5-Mev alpha particle which compares favorably with the best experimental value of 3 percent. For potassium iodide, whose light output is only 0.25 that of sodium iodide, the best resolution to be expected is about 5 percent, and this is roughly the experimental value obtained. These resolutions are usually sufficient to separate members of a decay chain, but, of course, are almost useless in looking for fine structure of a particular alpha emitter.

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3. Beta ray detector. --An anthracene crystal 1.5 inches in diameter by 1/4 inch thick was used for counting beta particles. It was mounted onto a Dumont-6292 phototube by means of the usual vaseline seal. In addition, to reduce light sensitivity a 0.0005-inch aluminum window was placed over the face of the crystal. Energy resolutions of 13 to 15 percent were obtained for the 0.63-Mev conversion line of Cs¹³⁷.

B. Coincidence Circuit

A block diagram of the coincidence circuit is shown in Fig. 1. The signal pulses are taken from the phototube through a preamplifier and linear amplifier and then branched. One branch is passed through a shaper, which gives the pulse a length of about 7×10^{-6} second, and then after passing through a flattener and another amplifier it is fed into the signal side of the 50-channel pulse-height analyzer. These pulses can appear on the registers, however, only when a simultaneous gate is received by the analyzer. The other branch of the signal pulses is passed into channel one of a variable gate-variable delay unit which is used to shape this pulse to about 0.3×10^{-6} second. Also a variable delay up to 10×10^{-6} second can be introduced. The output from this unit is sent into a coincidence mixer, from which point several possible alternatives will be discussed presently. The pulses from the gate phototube are passed through a preamplifier and then into a single-channel analyzer. Here, by means of a discriminator circuit pulses of any particular

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Fig. 1. Block diagram of the coincidence circuit.

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height may be selected. A variable window width also affords the possibility of accepting pulses over a longer interval. After passing through a linear amplifier the selected pulses are fed into channel two of the variable gate-variable delay unit. Again the pulse is shaped to about 0.3×10^{-6} second, and then sent into the coincidence mixer. In this unit a switching system makes it possible to select pulses from either separate channel or coincidence pulses between the two channels. The desired pulses are then reshaped to about 1×10^{-6} second and fed into the gate side of the 50-channel analyzer. The switching system on the coincidence mixer is quite convenient, as it makes it possible to run singles spectra by selecting the pulse branched from the signal side of the circuit. This is advantageous for energy calibrations as well as detailed comparisons between singles and coincidence spectra. When the gate pulses are selected, coincidences are run at the 50-channel analyzer with a resolving time of about 8×10^{-6} second. This arrangement has the advantage that the efficiency of the coincidence circuit is 100 percent, and by counting the gate pulses separately and measuring the signal geometry, the intensity of coincident events may be calculated. On the other hand when the coincidence pulses are selected from the mixer, the resolving time drops to around 0.6×10^{-6} second, but the efficiency also drops to 50 to 75 percent. Although not useful for intensity measurements, this arrangement is essential when high counting rates are encountered, and the chance coincidence rate, which is proportional to resolving time, would be too large with the slower set-up.

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A chance coincidence occurs when two unrelated pulses happen to strike within the resolving time of a coincidence circuit. A good approximation of the chance coincidence rate is given by the equation, $C = \tau \alpha \beta$, where C is the chance rate, τ the resolving time of the instrument and α and β the counting rates in the two single channels. In the present work it was not possible to count the pulses in each of the 50 signal channels, so that the above equation could not conveniently be applied. The chance rate was therefore experimentally determined by using an external unrelated source of gate pulses, the number of which was normalized to the number of gates generated during the actual experiment. If there was decay of the signal events during a run, it was, of course, necessary to make suitable corrections in the normalization. All the coincidence spectra discussed will have been corrected in this manner.

C. Angular Correlations

The radiations emitted from a radioactive source will, under normal conditions, always be isotropic in space, that is their intensity will be independent of the direction in which it is measured. However, when two radiations are successively emitted from a nucleus, it is possible by observing the first radiation in a particular direction to establish a group of nuclei with a preferred spacial orientation. The second radiations from these nuclei will not in general be isotropic, providing the nucleus is not reoriented during the time between the two emissions. Coincidence counting makes it possible to observe

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the second radiation only from the oriented nuclei, and a directional anisotropy or angular correlation may be observed. The shape of such correlations gives information regarding the types of radiations involved and also about the spins of the initial, intermediate, and final levels. Several angular correlations have been measured, the general procedure being to fix one counter and examine the angular distribution of coincidences by rotating the other one. Corrections, of course, should be made for the sample size and for the finite acceptance angle of the detectors, both of which are assumed to be negligibly small in the theoretical calculation of angular correlations. Individual techniques and corrections will be discussed as they arise.

D. Alpha Particle Spectrograph

The alpha particle spectrograph used in a number of experiments has been described elsewhere.⁸ It employs a 60° sector magnet and the normal trajectory has a radius of curvature of 75 cm. The transmission and energy resolution depend on the slit system used, but for normal operation have values around 4×10^{-4} percent and 0.1 percent, respectively. The receiver consists of a photographic plate from which the alpha particle tracks are individually counted under a microscope. Very thin samples were prepared by vacuum sublimation from tungsten filaments.

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III. EXPERIMENTAL RESULTS

A. Even-Even Nuclei

As implied earlier the study of even-even nuclei was divided roughly into two parts. The first part was concerned with extending the region over which information is available on the well-known rotational states.^{9,10} Included in this group were studies of 100^{254} , $\text{Cf}^{250-252}$, and Cf^{246} . The second part consisted of a detailed examination of several isotopes in the radium-thorium region. The interest here was in connection with the rather unexpected low-lying 1- states found. The data obtained will be discussed and compared with those for other even-even isotopes in the section on conclusions.

1. 100^{254} decay. --The isotope 100^{254} is prepared by the intensive neutron irradiation of lower elements, ultimately uranium.¹¹⁻¹⁶ Of interest here is the fact that californium isotopes, arising from β^- decay of lower elements, capture neutrons successively until the 20-day β^- -emitter, Cf^{253} , is reached. This decays to 99^{253} which is an alpha emitter of 20-day half-life which captures a neutron to give the 36-hour β^- -emitter 99^{254} . The 3-hour alpha-emitter 100^{254} soon grows into equilibrium with its parent. In order to take advantage of the longer half-life of 99^{254} in working on 100^{254} , chemical isolation of the 99 fraction was effected. Such preparations therefore contained:

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(1) 99^{253}	20-day half-life	6.64-Mev α
(2) 99^{254}	36-hour half-life	β^-
(3) 99^{254}	>1-year half-life	6.44-Mev α (very low)
(4) 100^{254}	3.2-hour half-life	7.22-Mev α

For the present measurements three preparations were employed. The first of these was of low intensity and consisted of 7000 disintegrations per minute of 99^{253} and 300 disintegrations per minute of 36-hour 99^{254} in equilibrium with its daughter, 100^{254} . The sample was mounted on an aluminum plate 0.0005-inch thick. The second and third samples were more intense and the 100^{254} had been separated chemically from element 99 and was essentially isotopically pure. At the start of measurements each of these two samples contained 3×10^4 disintegrations per minute and they were mounted on 0.0005-inch platinum foil. No sample of sufficient intensity for the alpha particle spectrograph has yet been available.

The weak source (containing mixed radioactivities) was the first one available and was used to establish the population of alpha transitions to the first excited state (see Fig. 3). On the assumption that the decay scheme of 100^{254} is not unlike that of other even-even nuclei such as $\text{Cf}^{246, 17}$, $\text{Cm}^{242, 18}$ and $\text{Pu}^{238, 19}$ this transition should lead to a 2+ state some 40 kev above the ground state which would be de-excited by a highly converted gamma transition. The measurement of the L x-ray intensity would therefore form the basis for calculating the population of the 2+ state.

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The alpha particle-L x-ray coincidence rate was determined in the following manner. The gamma geometry of the arrangement was calibrated using the 60-kev gamma ray of Am^{241} and taking its intensity to be 0.40 per alpha particle.²⁰ The absorption and fluorescence yield²¹ corrections were made as second-order corrections by comparison in the same apparatus with the somewhat softer L x-rays from the decay of Cm^{242} , in which case the alpha population to the 2+ level is known to be 26.3 percent.¹⁸ The alpha particles of 100^{254} were separated from those of 99^{253} by using a potassium iodide crystal. The resolution of this crystal at the high geometry used was 7.6 percent, which was barely enough to exclude 99^{253} alpha pulses when the gate single-channel analyzer was set to accept those from 100^{254} . The final corrected intensity to the 2+ state was 15 ± 2 percent.

The second and third preparations were used for an examination of the gamma ray spectrum. Coincidence counting was again employed in order to decrease background effects, but since alpha energy discrimination was not necessary, a zinc sulfide screen was used for alpha particle detection. The composite data for each sample are shown in Fig. 2a and 2b. Each of these curves is composed of several shorter runs which were made in order to be certain the coincidences decreased with the three-hour half-life of 100^{254} . In both instances this was found to be the case. The curve for preparation two (Fig. 2a) showed gamma rays of 42 ± 4 kev and 94 ± 2 kev. The peak at 66 kev is seen when any emitter of gamma rays of

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Fig. 2a. Alpha particle-gamma ray coincidence spectrum of 100^{254} from preparation two.

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sufficient energy is counted on platinum and is due principally to K x-rays of platinum produced by fluorescence excitation. There may be small contributions at this energy from the escape peak of the 94-kev gamma ray and also from backscattered radiation. The line showing the "maximum contribution from spontaneous fission" was determined in the following manner. The isotope Cf^{252} which has a high ratio of spontaneous fission to alpha disintegration^{22, 23} had been used to determine the gamma ray spectrum in coincidence with the fission process as it registers in the detection system employed here. The gamma ray intensity as a function of energy in the range of interest here was flat. It was assumed that this distribution would be the same for 100^{254} and the calculated intensity is shown in Fig. 2a as a maximum value since all of the spontaneous fission fragments may not have been counted by the zinc sulfide detector. In calculating the intensities of the gamma rays from the alpha decay process a spontaneous fission gamma ray background of one-half the maximum value shown was assumed. This assumption could introduce considerable error in the intensity of the 94-kev gamma ray and a correspondingly larger error for the 42-kev peak. From the third preparation (Fig. 2b) the same gamma rays were observed, however the statistics were considerably better and the spontaneous fission background much more clearly defined. The gamma ray energies observed were 41 ± 2 kev and 97 ± 2 kev.

The best energy for the lower energy gamma ray was taken to be 41 ± 2 kev. Its intensity was 2×10^{-2} percent (of the alpha

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disintegrations) and under the very likely assumption that the gamma ray is from the transition giving rise to the L x-rays, the conversion coefficient can be calculated. It will be recalled that L x-ray measurements indicated 15 percent population of the first excited state; therefore the conversion coefficient is 750. Within the accuracy of our information this is in good agreement with an E2 transition similar to that found for all other even-even nuclei in this region.

Probably the best energy for the other gamma ray is 96 ± 2 kev. This transition also has its analog in the alpha decay of other heavy even-even nuclei and has been placed accordingly in the decay scheme (Fig. 3) defining a state at 138 kev. The energy spacing agrees with the assignment to a 4+ state as part of a rotational band according to the Bohr-Mottelson theory.^{9,10} The intensity of this gamma ray was 4.4×10^{-2} percent from the first sample, but only 2.8×10^{-2} percent from the second. The latter figure is taken as the more reliable due to the spontaneous fission background uncertainty in the former. If the conversion coefficient of this gamma ray is taken to be the same as for the 103-kev transition of Cm^{242} (and this should represent a minimum) then the alpha population to the 138-kev level turns out to be at least 0.2 percent. From this a maximum hindrance factor of 150 may be calculated for this alpha group.

2. $\text{Cf}^{250-252}$ decay. --Mixtures of Cf^{250} and Cf^{252} can be produced by intensive neutron irradiation of lower elements,^{22, 23} and,

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Fig. 3. Decay scheme of 100^{254} .

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in fact, their production is a step toward the already described preparation of 100^{254} . The pertinent reactions in this case are as follows:

The isotopes listed are beta-stable except those designated as β^- emitters. Of the alpha activity in the sample, about 85 percent was due to Cf^{252} and 15 percent to Cf^{250} . The Cf^{249} could be estimated from the irradiation history to contribute only about 0.04 percent to the alpha activity. (The mass of Cf^{249} is only about ten fold lower than that of Cf^{250} , but its half-life is 400 to 500 years, whereas the half-life for Cf^{250} is only 10 years.) Despite the low activity of Cf^{249} , it has prominent gamma rays which showed up in the gamma ray spectrum as will be mentioned. Similarly the Cf^{251} can be estimated to be in even lower intensity. There should have been a relatively large amount of activity accompanying the beta decay of Cf^{253} , but since its alpha branching must be extremely small and the gamma rays were measured by alpha-gamma coincidence counting, this isotope should have been inert as far as the present measurements are concerned.

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After chemical purification of the californium fraction, a part of the activity consisting of 1.2×10^5 alpha disintegrations per minute was electroplated onto a 0.0005-inch platinum plate.²⁴ This plate was used as the alpha particle spectrograph source. The alpha-gamma coincidence source was prepared by evaporating to dryness a few drops of lactic acid solution containing 1.4×10^5 alpha disintegrations per minute on a 0.0005-inch platinum plate. The alpha-L x-ray source was prepared by evaporating to dryness on a 0.0005-inch aluminum plate a few drops of concentrated nitric acid containing about 2000 alpha disintegrations per minute.

On the assumption, which will later be shown to be justified, that Cf²⁵² and Cf²⁵⁰ have decay characteristics very much like other even-even nuclei in this region, the alpha population to the first excited state (around 40 kev) may be obtained by a quantitative measurement of L x-ray-alpha coincidences. This, of course, is due to the fact that the gamma transition to the ground state will be heavily converted in the L shell, and all other sources of L x-rays are very small compared to this one. Using exactly the same procedure as outlined for 100²⁵⁴, the alpha-L x-ray coincidence rate was compared with that of Cm²⁴², whose branching to the analogous level is known, and second-order corrections for fluorescence yields and for absorption were applied. It should here be mentioned that since the zinc sulfide screen was used for detecting alpha particles, fission fragments were also counted, and therefore spontaneous fission-gamma ray coincidences could be observed. However, these were small compared with the alpha-L x-ray peak. The number obtained

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for the population of the first excited states was 15.5 percent, which represents a weighted average of the values for Cf^{252} and Cf^{250} . It will be of interest to compare this number with the one obtained from the alpha particle spectrum.

Four measurements of the alpha particle-gamma ray coincidence spectrum were made. In this case the potassium iodide crystal alpha detector was used, and the alpha peak was quite easily resolved from the peak due to spontaneous fission fragments. It was considered possible, however, that some fission fragments which had been degraded in energy might give rise to pulses of a comparable height with alpha pulses and therefore the pure spontaneous fission-gamma ray coincidence spectrum was taken for comparison purposes. The energies and abundances of the radiations observed in coincidence with alpha particles are shown in Table I. A graph of the results of Experiment 2 is shown in Fig. 4. In coincidence with fission fragments, a gamma ray "continuum" was observed which rose to a plateau at 125 → 200 kev and then decreased continuously out to several Mev. Superimposed on the continuum was a peak at approximately 63 kev. This may well have been the platinum K x-rays produced in the sample backing plate by the fission gamma rays. Figure 5 shows the fission fragment-photon coincidences in the region of interest. From this spectrum it seems unlikely that any of the gamma rays shown in Table I are a result of the spontaneous fission process.

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Table I

Gamma Rays Observed in Cf²⁵⁰⁻²⁵² Sample

Exp. No.	42-kev gamma ray		67-kev radiation		100-kev gamma ray		K x-ray maximum ~180-kev gamma ray		Other radiation		
	Energy	Abundance	Energy	Abundance	Energy	Abundance	abundance	Energy	Abundance	Energy	Abundance
	(kev)	(x10 ⁻⁴)	(kev)		(kev)	(x10 ⁻⁴)	(x10 ⁻⁵)	(kev)		(kev)	(x10 ⁻⁴)
1	43	1.5	66-73	2.6x10 ⁻⁵	99	1.3	5	~190	>8x10 ⁻⁵ *	**	--
2	41	1.3	67	4.3x10 ⁻⁵	96	1.0	7	--	--	**	--
3	~40	1.5	70	1.2x10 ⁻⁴	99	1.4	9	~180	2.2x10 ⁻⁴	**	--
4	~40	1.4	63-67	2.5x10 ⁻⁵	100	1.4	7	~180	8x10 ⁻⁵	350-400	3

Best value

42	1.4	--	--	100	1.3	--	--	--	350-400	3
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*This gamma ray peak was only partially observed in this experiment.

**These gamma rays would not have been observed in this experiment.

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Fig. 4. Alpha particle-gamma ray coincidence spectrum of $\text{Cf}^{250-252}$.

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Fig. 5. Spontaneous fission fragment-gamma ray coincidence spectrum of Cf²⁵⁰⁻²⁵².

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The alpha spectra of Cf^{250} and Cf^{252} were observed,²⁵ using the alpha particle spectrograph. The energies and abundances of the groups found were 6.112 ± 0.005 Mev (84.2 percent), and 6.068 Mev (15.8 percent) for Cf^{252} and 6.024 ± 0.005 Mev (82.5 percent) and 5.980 Mev (17.5 percent) for Cf^{250} . This work is in good agreement with the previously reported energies.^{22, 23} The sample was found to contain 85.8 percent Cf^{252} and 14.2 percent Cf^{250} , so that the weighted average of the population to the first excited state is 16.0 percent. This is in excellent agreement with the 15.5 percent found from alpha-L x-ray coincidences, and therefore lends some confidence to the coincidence method.

The "42-kev" gamma ray is interpreted as belonging to the highly converted transitions of 44 kev (alpha particle energy difference plus recoil energy difference) leading from the first excited states to the ground states of both Cf^{250} and Cf^{252} . The energy measurement from the alpha data is somewhat better than the gamma ray value, so 44 kev is preferred. The abundance of this gamma ray is 1.4×10^{-4} per alpha particle (Table I). With the weighted abundance of the alpha groups to the 44-kev levels of 16 percent, the conversion coefficient is 1100. Since the sample activity is predominantly Cf^{252} , this conversion coefficient applies principally to Cf^{252} . The fact that the analogous transition in Cf^{246} decay has a conversion coefficient of about this same value, makes it seem quite reasonable that the amount of Cf^{250} present will not affect this number greatly. By comparing this conversion coefficient with theoretical values, it

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is found that the transition can only be E2. Since the ground state of Cm²⁴⁸ (daughter of Cf²⁵²) is assumed to have spin zero, even parity, this requires that the first excited state have spin two, even parity. This is, of course, expected since nearly all even-even nuclei are found to have a 2+ first excited state.

Whenever a photon of approximately 100 kev is seen in the heavy-element region it may be a K x-ray or a gamma ray or both. For curium the K x-ray group should produce a peak of approximately 108 kev, and it is seen from Fig. 4 that most of the peak at approximately 100 kev lies at too low an energy. However, the peak does tail on the high-energy side and it seems likely that it is not produced by a single gamma ray. In order to get a minimum value for the intensity of the gamma ray, a peak centering on the K x-ray energy of 108 kev was resolved. The shape of the subtracted peak was obtained by weighting the different components of the K x-ray group according to the relative intensities observed for uranium. The best energy for the residual peak is 99 kev; it would be 101 if the K x-ray subtraction were not made. The minimum intensity of this gamma ray was 1.3×10^{-2} percent of the total alpha particles. The group attributed to K x-rays has a corresponding maximum intensity of 7×10^{-3} percent.

This gamma ray probably represents a transition from the second excited state to the first excited state of each of the isotopes, Cf²⁵² and Cf²⁵⁰. This assignment is made by analogy to other even-even alpha emitters in this region. Because of the preponderance of

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Cf^{252} in the sample, the measured energy and abundance of the gamma ray apply principally to Cf^{252} decay. Since these values are much the same as found in Cf^{246} decay, however, Cf^{250} would be expected to show in its decay a gamma ray of similar energy and abundance. The decay schemes of Cf^{250} and Cf^{252} consistent with all the data discussed are shown in Fig. 6.

Radiations whose energies were roughly 67, 180, and 350 to 400 kev were also observed. The 67-kev peak is probably due largely to K x-rays of platinum from fluorescence excitation of the backing plate. A sample of highly enriched Cf^{249} showed gamma rays of approximately 350 and 400 kev in high intensity, and this corresponds well with the highest energy radiations seen here. This decay will be discussed in a later section. The gamma ray at 180 kev is not accounted for by Cf^{249} , but could possibly come from Cf^{251} or, though less likely, from Cf^{253} .

3. Cf^{246} decay. -- The preparation of Cf^{246} in a state of high radioactive purity can be effected by irradiating a mixture of curium isotopes (Cm^{242-6}) with alpha particles of about 40-Mev energy.²⁶ (The mixture of curium isotopes is obtained by prolonged neutron irradiation of Pu^{239} .) Of the californium isotopes produced, Cf^{246} has a half-life of 35.7 hours,²⁶ while Cf^{247} , Cf^{244} , and presumably Cf^{245} have half-lives sufficiently short^{26, 27} to allow their complete decay during a period in which the Cf^{246} is not greatly reduced. No appreciable contribution to the radioactivity comes from Cf^{248}

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Fig. 6. Decay schemes of Cf^{250} and Cf^{252} .

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because of its relatively long half-life (250 days)²⁸ and low yield since there were only small amounts of Cm^{246} and Cm^{245} in the bombarded curium. Cm^{242} would be present as the daughter product of Cf^{246} decay.

The sample of Cf^{246} made available for the present study contained initially 2.9×10^5 alpha disintegrations per minute. There was some Cf^{250} and Cf^{252} in the sample due to their incomplete removal from the target curium before bombardment. Three preparations were made from the above sample. Roughly 10 percent of the total activity was evaporated to dryness on a 0.006-inch aluminum plate. This was used for alpha-L x-ray coincidences. The other 90 percent was prepared by vacuum sublimation and exposed in the alpha particle spectrograph for several days. After the exposure the activity was transferred to a 0.006-inch aluminum plate and used for alpha-gamma coincidences.

Once again the alpha-L x-ray coincidences were measured quantitatively in order to determine the branching to the first excited state of Cm^{242} (daughter of Cf^{246}). The reasoning and procedure are exactly the same as has been discussed for 100^{254} and $\text{Cf}^{250, 252}$ decay. The results indicate that 22 percent of the alpha transitions lead to the first excited state of Cm^{242} .

The gamma ray spectrum of Cf^{246} was observed in coincidence with alpha particles in order to reduce the background radiation. Some spontaneous fission-gamma ray coincidences were observed due to the Cf^{250} and Cf^{252} present in the sample, but this has been shown

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previously to contribute only an essentially uniform background. Four measurements of this spectrum were made over a period of almost two half-lives of Cf^{246} . The second of these measurements is shown in Fig. 7. In addition to L x-rays, peaks at roughly 44, 60, and 100 kev are seen, which, except for the one at 60 kev, decayed with the 36-hour Cf^{246} half-life. The 60-kev peak is probably due to Am^{241} impurity. Because of the high intensity of the Am^{241} 60-kev gamma ray and the low intensities of the Cf^{246} photons, an extremely small amount of Am^{241} (80 disintegrations per minute) would give the observed intensity.

Two exposures (22.6-hour and 33-hour, respectively) were made on the alpha particle spectrograph¹⁷ before the sample became too weak. In the first, 5137 tracks were registered: 78 percent in the main group and 22 percent in the group leading to the first excited state. This is in excellent agreement with the alpha-L x-ray coincidence value, which is also 22 percent. The energy difference between the two groups was found to be 42.0 kev. When correction is made for the differences in recoil energy, the corresponding gamma ray transition becomes 42.7 kev. The appearance of the first excited state of Cm^{242} is also prominent in the beta decay of $\text{Am}^{242\text{m}}$. The transition energy has been measured as 42.3 ± 0.2 kev with a bent crystal photon spectrometer (Jaffe),²⁹ and from the conversion electron spectrum, the value 42.2 ± 0.3 kev was obtained (Church).³⁰ This photon is also seen in the alpha-gamma coincidence spectrum of Cf^{246} (see Fig. 7), but the energy determination is not as accurate as the three values cited here.

Fig. 7. Alpha particle-gamma ray coincidence spectrum of Cf^{246} .

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In the second alpha spectrum taken, the photographic plate was also exposed to a source containing the U^{230} series. The Em^{218} line at 7.127 Mev³¹ was taken as the energy standard and from this, the main group of Cf^{246} was found to be 6.753 Mev. When the recoil energy correction is made, the decay energy for Cf^{246} becomes 6.865 Mev.

Because of the small number of events recorded a good intensity of the 42 (44) kev gamma ray could not be obtained. However, a value of roughly $(1.8 \pm 0.5) \times 10^{-4}$ per alpha disintegration was indicated. This, together with the population of 42-kev level (22 percent), would lead to a conversion coefficient roughly between 1000 and 1500. These values are in the expected range of an E2 transition, and, as would be expected, spin 2, even parity is indicated for the first excited state of Cm^{242} .

The average value for the intensity of the radiation around 100 kev was 1.4×10^{-4} photon per alpha disintegration. Since the energy of this photon corresponds well with the expected transition from the 4+ state to the 2+ state, we can assume that it is an E2 transition. Then if we assume that the conversion coefficient is the same as that measured for the corresponding transition in the decay of Cm^{242} ,¹⁸ the minimum intensity of population of the 4+ state becomes approximately 0.08 percent. This leads to a maximum hindrance factor for alpha decay to this level of 220.

The decay scheme of Cf^{246} constructed from these data is shown in Fig. 8.

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Fig. 8. Decay scheme of Cf^{246} .

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4. Ra²²⁴ and Th²²⁸ decay. --The Th²²⁸ and Ra²²⁴ were obtained from a mesothorium source of approximately 70 mg radium gamma ray equivalent. It will be seen that six alpha groups so far found are ascribable to these two isotopes and since the energies are close to each other, fairly rigorous chemical separations are required to assign the groups, particularly those in low intensity. Furthermore, possible interfering groups are at hand from the RaC (Bi²¹²) of this decay series as well as from members of the radium (Ra²²⁶) series present as minor contaminants in the mesothorium. The problem of analysis hinges on the growth rates of the members of the series. Th²²⁸ (1.9-year half-life) can be obtained pure initially but Ra²²⁴ grows in with its 3.64-day half-life. Other members rapidly follow the Ra²²⁴ growth except Bi²¹² (60-minute half-life) which is held up slightly by the growth of its beta-emitting parent, 10.6-hour Pb²¹². As may be visualized from these lifetimes, it is possible to make an alpha spectrum analysis of Th²²⁸ relatively free of decay products and of Ra²²⁴ with its products free of Th²²⁸, but it is not possible to analyze Ra²²⁴ free of its decay products. If Ra²²⁶ is present in the mesothorium, its decay products, Em²²² and Po²¹⁰, will produce alpha groups in the energy range of interest; Bi²¹⁴ is excluded from consideration because of its extremely low alpha branching.

The method of chemical separation made use of Dowex-50 ion - exchange resin upon which the radium and thorium fractions were adsorbed and eluted selectively. Samples were prepared whose alpha

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activity ranged from essentially pure Ra^{224} to 75 percent Th^{228} . This permitted the assignment of low intensity groups to be made by comparing among the various samples their abundance relative to the main peaks of the alpha emitters present. The details of these comparisons are contained in a previous paper.³²

The standard of reference for energy was the principal alpha group (α_0)* of Ra^{224} which had been determined as 5.681 Mev by Briggs.³³ In some instances energies were determined more conveniently relative to Em^{222} (radon) and Po^{210} after these were identified in the samples. Also other abundant groups of Ra^{224} and Th^{228} served as secondary standards.

4a. Ra^{224} . --The main alpha group of Ra^{224} (5.681 Mev) leading to the ground state of Em^{220} is readily identified and, as mentioned, has served as the energy standard for the present series of measurements. Rosenblum, Valadares, Perey, and Vial³⁴ have found an alpha group of Ra^{224} at 5.448 Mev in 4.6 percent abundance. This same group was observed in the present measurements. Its energy was determined to be 5.445 Mev and its abundance (of total Ra^{224} alpha particles) was 4.9 percent; both values are in agreement with those of Rosenblum and co-workers.³⁴

* α_x is used throughout to indicate the alpha group populating an excited state of energy x.

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The energy difference between the two alpha groups of Ra^{224} as found here is 236 kev, therefore the second group leads to an excited state of 240 kev. No measurement of the gamma ray transition was made here, but Rosenblum, Valadares, and Guillot³⁵ have observed K and L conversion lines of a 241-kev gamma ray and from the conversion coefficient deduced that the transition is of the E2 type. This conclusion designates the 240-kev state as 2+.

From the discussion of the Th^{228} spectrum which follows, it will be seen that no other alpha groups of Ra^{224} could be identified down to energies 884 kev below the main alpha group. This observation does not agree with the work of others.³⁴ On the basis of these measurements, the alpha decay scheme of Ra^{224} consists simply of the ground state transition in 95 percent abundance and the transition to the 240-kev 2+ state in 5 percent abundance. Any other alpha group within the range of these measurements is in <0.1 percent abundance.

4b. Th^{228} . --The principal alpha group of Th^{228} was measured as 5.421 ± 0.001 Mev, using $\text{Ra}^{224} \alpha_0$ as the standard. In the same experiment, the group leading to the first excited state was observed and its energy was found to be $5.388_5 \pm 0.001$ Mev and its abundance 28 percent. The energy difference defines the first excited state at 84.3 kev. In another run the abundance of the group was found to be 26 percent and the energy level 84.9 kev. The data on the two principal groups of Th^{228} are in excellent agreement with those of

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Rosenblum, Valadares, and Perey³⁶ who reported energies of 5.423 and 5.338 Mev in this same relative abundance.

Besides these two high abundance groups, two others in low intensity were observed which we attribute to Th^{228} . Figure 9 shows one spectrum taken covering the energy range from 5.15 to 5.71 Mev. The solid curve applies to the 19-hour exposure and on this the groups α_0 and α_{84} of Th^{228} and α_0 of Ra^{224} were too intense to be shown on the same scale. The peaks for these groups, shown as broken lines in Fig. 9, were obtained by making partial scans of the photographic plate and represent about 2.4 percent of the total tracks registered. It will be noted that groups due to Po^{210} and Em^{222} are on the plate. Other exposures on samples from which Po^{210} was absent and Em^{222} present in altered abundance have confirmed their assignments. An alpha group of Bi^{212} (5.603 Mev) falls within the energy range of these measurements but its estimated intensity would have been below our limit of detection.

The two low-intensity groups at 5.173 and 5.208 Mev bear constant relationship to the Th^{228} content of the sample and not to the amount of Ra^{224} . This observation is somewhat at variance with the results of Rosenblum and co-workers³⁴ who reported no groups of Th^{228} in this energy range but one at 5.194 Mev with Ra^{224} in 0.4 percent abundance. It will be seen that these two groups of Th^{228} fit in well with a proposed decay scheme which includes the gamma ray transitions. Aside from the Th^{228} groups already mentioned, an upper limit of 0.04 percent could be set for any in the energy range 4.80 to 5.67 Mev.

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Fig. 9. Alpha spectra of Th^{228} and Ra^{224} .

— Full scan across the width of the photographic plate. --- About 2.4 percent of full scan.

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In order to examine the gamma rays of Th^{228} free of Ra^{224} and its decay products, a quick chemical separation was devised. The mixture in dilute nitric acid solution was placed on a Dowex-50 resin column jacketed to allow operation at an elevated temperature (in this case 87°C) which permits rapid equilibration. The radium, lead, and bismuth fractions were eluted with 4 N nitric acid after which the thorium was stripped with a 50-volume percent solution of lactic acid at pH 3. The thorium solution was evaporated to dryness, the alpha activity measured, and then the gamma rays were analyzed with a scintillation counter coupled to a 50-channel pulse analyzer. The gamma ray spectrum was measured within one hour of the time that the decay products of Th^{228} were separated.

Figure 10 shows the gamma ray spectrum of Th^{228} in which is found a prominent peak at 89 kev and lesser peaks at 137, 169, and 212 kev. The assignment of these gamma rays to Th^{228} seems certain since the growth of the 238-kev gamma ray of Pb^{212} and those of Bi^{212} were readily followed and extrapolation of their abundances to time of initial measurement showed that separation of the decay products of Th^{228} was complete. The small peak on the low-energy side of the 89-kev peak is probably its escape peak. K x-rays from the internal conversion of the higher-energy gamma rays will be under the 89-kev peak. For calibrating the instrument, the 60-kev gamma ray of Am^{241} and the 184-kev gamma ray of U^{235} were employed. The standards could not be run concurrently with the Th^{228} ; as a result there are uncertainties in energy due to

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Fig. 10. Gamma ray spectrum of Th^{228} .

— Gamma ray counting rate indicated by ordinate scale. --- Gamma ray counting rate equals 10 times the value indicated by the ordinate scale.

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drift of photomultiplier voltage which has been found in practice to be less than one channel corresponding to 8 kev. Probably the gamma energies are good to about 5 kev.

The intensities of the gamma rays were determined by correcting the observed intensities for counter efficiency and for the escape peaks. Later data on these corrections have changed the intensities slightly from those given in a previous paper.³² The corrected values as percentages of the total Th^{228} alpha particles are shown in the decay scheme, Fig. 11. The energy levels shown are those determined from the alpha spectrum while the measured gamma ray energies are indicated along with their abundances by the vertical arrows designating the transitions.

The 89-kev gamma ray is almost surely the same as that assigned to a pair of close-lying gamma rays³⁷ or more recently to a single gamma ray of 84 kev³⁸ or 83 kev.³⁹ The energy of this state from our alpha decay data is 84 kev, which is closer to the above cited values^{38, 39} for the gamma ray energy. Riou³⁹ measured the intensity of the gamma ray (relative to total alpha particles) as 2.1 percent and relating this to the 28 percent abundance of α_{84} the conversion coefficient was found to be twelve.⁴⁰ From our measurements (1.7 percent intensity and 28 percent population by α_{84}) the conversion coefficient is 15. These conversion coefficients as well as the $L_I/L_{II}/L_{III}$ ratios correspond with the assignment to E2 radiation (from calculated conversion coefficients of Gellman, Griffith, and Stanley)⁴¹ as already pointed out by Rosenblum, Valadares

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Fig. 11. Decay scheme for Th^{228} .

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and Buillot.³⁸ Attempts have been made^{42, 43, 44} to assign the radiation uniquely by alpha-gamma angular correlations but although there is strong evidence^{43, 44} for an electric quadrupole component the correlation is not exact. Nevertheless, the preponderance of evidence in this case, as well as for even-even nuclei in general, points to 2+ for the first-excited state.

We shall consider next the 169-kev gamma ray. This energy agrees well with the difference in energy between the states populated by α_{253} and α_{84} . From the population of the 253-kev state (0.2 percent) and the intensity of the 169-kev gamma ray (0.12 percent) a total conversion coefficient of 0.7 is calculated. The sum of K^{45} and L^{41} theoretical conversion coefficients for E1, E2, and M1 radiation are estimated respectively to be 0.1, 1.2, and 4.9. The radiation is therefore probably E2 and the spectroscopic state designation is 0+, 2+, or 4+. Because of the absence of the cross-over transition and the apparent absence of any M1 admixture, the most probably designation is 4+, although 0+ is not ruled out from these considerations alone.

The picture so far discussed for Th^{228} corresponds precisely with the decay of another even-even nuclide, Cm^{242} .¹⁸ In the case of the excited states of Pu^{238} from alpha decay of Cm^{242} and beta decay of Np^{238} , it was deduced that the low-lying states have even parity and that no observed odd parity state lies lower than at least about 1 Mev. To explain the other gamma rays found in Th^{228} decay we are assuming that the first odd parity state of Ra^{224} has

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decreased in energy so that it lies among the even parity states according to the following arguments.

The energies of the 137-kev and 212-kev gamma rays agree within experimental error with transitions from the state populated by α_{217} to the first even-spin state and to the ground state. The sum of the gamma ray intensities is the same as the population of the 217-kev state by alpha particles within experimental error (actually larger by measurement) so that the conversion coefficients for both gamma rays must be considerably less than unity. This can only be fulfilled by assigning both to E1 transitions which makes the 217-kev level a 1- state.

There is additional evidence for the above conclusion. It happens that the theoretical directional correlation for a sequence involving an alpha particle transition between a 0+ and a 1- level followed by a gamma ray transition from the 1- level to a 0+ level is very marked.⁴⁶

This correlation is plotted in Fig. 12 and it is seen that there is a minimum at 180° between the alpha particle and gamma ray detectors, with corresponding maxima at 90° and 270° . Since the ground states of Th^{228} and Ra^{224} are expected to be 0+, the above situation should apply to the alpha particles populating the 217-kev level of Ra^{224} and the 217 (212) kev gamma ray if the 1- assignment is correct. That this appears to be the case is shown by the following experiment. A Th^{228} sample, separated from its decay products by the resin column technique, was simply evaporated from aqueous solution as a 1/4 inch diameter spot on a 0.006-inch thick aluminum plate. The low atomic

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Fig. 12. Theoretical alpha-gamma angular correlation for a $0^+ \xrightarrow{\alpha} 1^- \xrightarrow{\gamma} 0^+$ sequence.

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number was employed for the backing since the gamma rays had to penetrate it to reach the detector and it was desired to avoid different absorptions due to different angles of penetration.. This sample is considerably larger than is desirable for precise angular correlation measurements, but it will be seen that another factor prevented determination of the precise correlation, so that undue care here did not seem warranted.

The correlation obtained by gating with the alpha particles and rotating the gamma ray detector is shown in Fig. 13. The limits shown here are the statistical limits due to the number of counts accumulated. This number was small because the intensity of the 217-kev gamma ray is low, and the time available was limited by the growth of decay-product activity. It is seen that maxima and minima occur in the same positions as in the theoretical correlation but the angular anisotropy is not so pronounced for the experimental data. Two of the most important reasons for this are the large sample size and relatively high geometry, for which no corrections were made. Nevertheless this correlation may be taken as rather good support for the 1- assignment of the 217-kev state of Th^{228} , particularly since there are limitations in other possible assignments. From the alpha particle and gamma ray data just discussed it is fairly certain that the 217-kev gamma ray leads to the ground state of Ra^{224} and is therefore electric radiation. (The conservation of spin and parity demands that a gamma ray must be electric if it leads to the ground state of an even-even nucleus and arises from a

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Fig. 13. Alpha-gamma angular correlation for the 217-kev transition of Th^{228} decay.

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level populated by alpha decay.) Assignment of E3 and higher multipolarities can be ruled out by consideration of lifetimes since in this case prompt coincidences were observed (few microseconds maximum delay) whereas the calculated lifetime for a E3 transition would be 10^6 fold longer. Therefore, we need consider only E1 or E2 assignments and the angular correlation expected if the radiation were E2 (corresponding to a $2+$ level) is quite different from that observed.

All these data are consistent with the decay scheme shown in Fig. 11, which, with the possible exception of the $4+$ assignment of the 253-kev level, may be considered well confirmed.

5. Th²³⁰ decay. --An investigation of the alpha decay of Th²³⁰ was undertaken with the thought that due to its proximity to Th²²⁸ a $1-$ state might appear among the energy levels of the daughter, Ra²²⁶. There was, in fact, previous evidence that a level existed in Ra²²⁶ at around 250 kev,^{47, 48, 49} which was not a member of the usual rotational band. Since a satisfactory alpha particle spectrum has not yet been obtained on the spectrograph because of the low specific activity of Th²³⁰, a study of the gamma rays offers the best approach to this decay.

Highly enriched samples containing about 90 percent Th²³⁰ and 10 percent Th²³² were obtained. These samples had been enriched on the Oak Ridge mass separators, and were available in quantities of up to 200 mg. Chemical separations from the decay products of Th²³⁰

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(as well as from those of the Th^{232} , Th^{228} family present in very low abundance) were made using the resin column technique described for Th^{228} . This method had been found in the case of Th^{228} to reduce daughter activities beyond the limit of detection, and again this was found to be the case, even though the limit was quite low. Three samples ranging in activity from approximately 4×10^6 alpha disintegrations per minute (100 μg) to 10^8 alpha disintegrations per minute (2.5 mg) were purified in this manner.

The gamma ray spectrum of Th^{230} obtained with a sodium iodide (thallium activated) scintillation spectrometer is shown in Fig. 14. The energies and relative intensities of the observed radiations are shown in Table II. Also shown here are the corresponding data of other workers. Among the more accurate measurements the agreement is seen to be good except in the case of the 184-kev gamma ray. Several comments concerning this gamma ray are in order. First, if the energy resolution of the crystal were only slightly poorer, it would be impossible to separate this peak from the one at 142 kev. This is undoubtedly the reason it has not previously been identified. Secondly, this is almost surely not the gamma ray associated with the decay of the daughter, Ra^{226} , because no higher-energy radiations associated with other daughter activities were found to be present or to grow in with the 3.8-day half-life of Em^{222} . Finally, the energy cannot possibly be raised to around 200 kev in better agreement with other investigators due to the availability of a U^{235} energy standard, which has a peak only 1 kev away at 185 kev. It might also be mentioned here

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Fig. 14. Gamma-ray spectrum of Th^{230} .

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Table II
Gamma Rays of Th²³⁰

Investigator	Method	Absolute abundance L x-rays (%)	68-kev gamma Energy (kev)	Rel. Abund. *	142-kev gamma Energy (kev)	Rel. Abund.	184-kev gamma Energy (kev)	Rel. Abund.	253-kev gamma Energy (kev)	Rel. Abund.
I. Curie ⁵⁰	Abs.	11	68	100	140	38	--	--	240	6
M. Riou ⁵¹	Abs.	9±2	68±2	100	190±20	60	--	--	--	--
Bouissiers et al. ⁵²	Scint. spec.	--	67	100	150±3	12.5	200±15	~1	254±5	3
Rasetti and Booth ⁴⁹	Scint. spec.	11.7	68±1	100	142±2	13	--	--	255±3	4
Rosenblum et al. ⁵³	Elect. spec.	--	67.75	--	141	--	203	--	255	--
Present work	Scint. spec.	**	68±2	100	142±2	11.8	184±2	2.3	253±2	2.9
Absolute Inten. (in %/α dis.)			0.59		0.070		0.014		0.017	

* Normalized to 100.

** L x-rays would have been absorbed in the aluminum window of the detector.

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that the plateau between the 184- and 253-keV peaks in Fig. 14 is very probably due to the simultaneous arrival of a 68- and a 142-keV photon at the detector. These two gamma rays will later be shown to be in coincidence.

Although the relative intensity data shown in Table II are in reasonably good agreement, this is not the case with absolute intensity measurements made on the 68-keV photon which are shown in Table III.

Table III
Intensity of the 68-keV Gamma Ray of Th^{230}

Investigator	I. Curie ⁵⁰	Riou ⁵¹	Bouissiers et al. ⁵²	Rasetti and Booth ⁴⁹	Present work
Absolute intensity of 68-keV γ (In %/a dis.)	0.85	0.50 ± 0.15	0.33 ± 0.1	1.05 ± 0.1	0.59 ± 0.05

The determination from the present work was made on two samples, vacuum sublimed onto 0.002-inch platinum plates. Alpha particle counting was done in an argon proportional counter at 52 percent geometry. Both samples were shown to be quite thin as far as attenuation of the alpha particle energy was concerned. One sample contained 6.1×10^4 and the other 1.4×10^5 alpha disintegrations per minute, or about 1.5 and 3 μg , respectively. Alpha pulse-height analysis of the second sample showed no alpha groups except those

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attributed to Th^{230} . The 68-keV gamma ray was compared with the one at 60 keV in Am^{241} , in which case an intensity of 0.40 photon per alpha particle²⁰ was used. The Am^{241} had been alpha counted in the same way as the Th^{230} . The fact that the result given in Table III lies at about the average of all previous results was added encouragement for its use. The absolute gamma ray intensities based on 0.59 ± 0.05 percent 68-keV photon per alpha disintegration and the relative intensities from the present work are shown in the bottom row of Table II. A limit of 6×10^{-7} photon per alpha disintegration could be set on any other radiation between 300 and 700 keV.

Gamma-gamma coincidence studies were made on Th^{230} with the following results. In coincidence with the 68-keV peak, the 142- and 184-keV gamma rays appeared in roughly the same relative intensity they have in the gamma ray spectrum. Since the 68-keV gamma ray almost certainly represents the transition from the first excited state to the ground state, this probably indicates that both the above gamma rays proceed to the first excited state of Ra^{226} . The 253-keV gamma ray was not observed, the limit of detection being about a factor of 7 or 8 lower than the intensity of the 184-keV peak. In coincidence with the 142-keV gamma ray only the 68-keV peak showed up in appreciable intensity, and with the 253-keV gamma ray no significant radiations appeared. The final measurement was made in coincidence with L x-rays and here an interesting result was obtained. The L x-rays arise predominantly from the conversion of the 68-keV gamma ray, and therefore the coincident spectrum

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should resemble that obtained in coincidence with the 68-kev peak. This was indeed found to be the case in that again the 142- and 184-kev peaks appeared in roughly the same relative intensity observed in the gamma ray spectrum, but this time peaks were also seen at 68 kev (due to L x-ray gates from the conversion of the 142-kev gamma ray and to platinum K x-rays from the sample backing plate), at 86 kev (probably radium K x-rays which should be greatly enhanced), and at 253 kev. The 253-kev peak was about a factor of ten lower in intensity than the peak at 184 kev, and it appeared in this same relative intensity on each of four separate runs, one of which is shown in Fig. 15. A possible interpretation of this result will be discussed later.

Valladas and Bernas⁴⁷ have observed the alpha particle spectrum of Th^{230} using an argon ion chamber. They find alpha groups as follows: α_0 , α_0 -68, α_0 -210 $\pm$ 10 (about 0.12 percent) and α_0 -245 $\pm$ 15 (about 0.03 percent). Rosenblum et al.⁴⁸ have seen these same groups on their alpha particle spectrograph, and in addition several others of low intensity. Their abundances for α_0 and α_0 -68 kev are 76.3 and 23.4 percent, respectively. Although the group which they find corresponding to α_0 -210 kev (mentioned above) looks real, the rest of their low-intensity groups are so small compared with the background, that they will not be considered further.

These data, together with the gamma rays, the gamma-gamma coincidences and some angular correlation work⁵⁴ indicate the decay scheme shown in Fig. 16, the major features of which have been

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Fig. 15. Gamma ray spectrum of Th^{230} in coincidence with L x-rays.

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Fig. 16. Decay scheme of Th^{230} .

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previously suggested.⁴⁹ By conversion coefficient⁵¹ and angular correlation⁵⁴ arguments already made in detail elsewhere, the 68-keV gamma ray is very likely E2, making the first excited state 2+, as is found for almost all even-even nuclei. Because of the energy level spacing and the absence of a crossover transition to the ground state, it seems very probable that the 210-keV level is the third member of the rotational band, which is well characterized in this region. The angular correlation work,⁵⁴ although not conclusive, is most consistent with a spin of 4+ for this level, as is expected for the third rotational level in even-even nuclei. In line with this proposal is a rough value of 0.7 for the conversion coefficient of the 142-keV gamma ray, which is perhaps most consistent with an E2 transition, although others cannot be ruled out. On this basis a tentative assignment of 4+ is given to the 210-keV level.

The energy and decay characteristics of the 253-keV level are so similar to those of proven 1- states in this region (Th^{228} decay already discussed and Th^{226} and U^{230} decay which will be discussed), that a 1- assignment of this level is strongly suggested. This would imply that the 184- and 253-keV gamma rays were both E1, and although there is no conclusive evidence for this, there are two indications that the conversion coefficients are both rather small (as expected for E1 transitions). First, the sum of the intensity of the two gamma ray transitions is 0.03 percent, which when compared with the alpha population of 0.03 ± 0.015 percent measured by Valladas and Bernas⁴⁷ indicates a conversion coefficient less than 0.5. Then too Valladas

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and Bernas were unable to see electron-alpha coincidences with the alpha particles populating the 253-kev level, and they mention that this is indicative of a low conversion coefficient of the radiation from this level. However, because there is no conclusive evidence, the 1- assignment must also be considered tentative.

The final detail of the decay scheme shown in Fig. 16 is derived from the L x-ray-253-kev gamma ray coincidences. Since the 253-kev transition is believed to go to the ground state, the presence of such coincidences implies alpha population to a level higher in energy than 253 kev, which subsequently decays to the 253-kev level by a transition that is L (or possibly K) converted. Furthermore, by assuming each 184-kev photon is in coincidence with about one L vacancy (the 68-kev gamma ray is highly L converted), and comparing the relative intensity of the 184- and 253-kev peaks in the raw gamma spectrum with that in the L x-ray coincidence spectrum, it is possible to estimate that roughly 10 percent of the 253-kev gamma rays are in coincidence with L vacancies. Now, from coincidences with the 253-kev peak, a limit can be set on the intensity of any gamma ray which might give rise to the observed L vacancies, and this limit is such that the L-shell conversion coefficient must be at least the order of 150 if only one transition occurs. This suggests that the transition is of low energy. It should be emphasized that these last conclusions are of a speculative nature.

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6. Ra²²⁶ decay. --The occurrence of low-lying 1- levels in Ra²²⁴ and Ra²²⁶ from the alpha decay of Th²²⁸ and Th²³⁰ was of considerable interest, and prompted an investigation of as many even-even alpha emitters in this region as was feasible. It was for this reason the present measurements on Ra²²⁶ were made.

Large quantities of Ra²²⁶ from natural uranium were available. The samples which were used here were portions of larger ones which had been vacuum-sublimed onto 0.002-inch platinum plates. These larger samples had previously been used for examination of the alpha particle spectrum on the spectrograph.⁵⁵ The two portions used in the present study contained roughly 10^5 and 10^6 alpha disintegrations per minute.

The object of the experiment undertaken was a very careful examination of the gamma ray spectrum accompanying the alpha decay of Ra²²⁶. In order to do this it was necessary that gamma rays from other sources be reduced to an extremely low level. Alpha-gamma coincidences could effectively remove radiations not associated with alpha particle emitters, but the background level was still found to be high, due to the presence of the alpha-emitting daughter activities of Ra²²⁶. If, however, the alpha particle spectrum was pulse-height analyzed, and only those pulses between about 100 and 400 kev lower in energy than the Ra²²⁶ main peak were selected as gate pulses, then the background became about an order of magnitude lower and the experiment was possible. The pulse-height analysis was done with a potassium iodide (thallium activated) crystal as has been described,

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with which it was possible to resolve the various alpha emitters of the family almost completely. Figure 17 shows the alpha particle spectrum obtained with the potassium iodide crystal. A large additional lowering of background radiations was obtained by lightly flaming the sample to remove Em^{222} (and subsequently its daughters). This method could also be used to prove the observed gamma rays did not belong to daughter activities.

The gamma ray spectrum obtained in this manner is shown in Fig. 18. The peak at 188 kev is the well-known gamma ray of $\text{Ra}^{226, 56}$ and the lower energy peak is probably a mixture of emanation K x-rays from the conversion of the 188-kev photon and platinum K x-rays from the sample backing plate. Also seen is a low-intensity peak at 255 ± 5 kev. This peak appeared in about the same intensity on two other determinations, and when compared with the 188-kev peak, which was assumed to be present to an extent of 3.0 percent per alpha disintegration,^{40, 55} this intensity was $(1.0 \pm 0.2) \times 10^{-2}$ percent per alpha disintegration. When the alpha particles were detected using a zinc sulfide screen with no pulse-height selection, it was possible to see this peak above background only if the sample had been flamed. In this case its intensity relative to the 188-kev peak was very nearly the same as when the alpha pulses were selected as previously described. This makes it seem unlikely that the gamma ray is due to anything other than Ra^{226} or an alpha emitter of very nearly the same alpha particle energy.

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Fig. 17. Alpha particle spectrum of a Ra²²⁶ family taken with a potassium iodide crystal.

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Fig. 18. Alpha particle-gamma ray coincidence spectrum of Ra^{226} .

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The decay scheme suggested for Ra^{226} from previous data contains only one excited state at 188 kev. The 255-kev gamma ray could terminate at either the ground state or this 188-kev level; however the former is considered slightly more probable on the basis of the invariance of the relative intensities of the 188- and 255-kev peaks among the three runs made. If the alpha groups leading to these gamma rays were separated by 255 kev, this would seem to be less likely than if they were separated by only about 70 kev. A possible way to confirm this would be to look for a peak at about 70 kev in coincidence with the 188-kev gamma ray. This was tried without success, however a significantly low limit could not be set.

In conclusion it may be said that if the 255-kev level does exist it is of about the same energy as other 1- levels in this region.

7. U^{230} family decay. --The final group of even-even nuclei studied was the U^{230} family. Once again one of the primary interests was to extend our knowledge of the low-lying 1- states appearing in even-even nuclei in this region. As in the case of Th^{230} , there was previous evidence that unexplained levels existed in U^{230} and Th^{226} ³¹

The U^{230} was made by bombarding thorium metal with 100-Mev protons. The target was allowed to stand about a month to give the Pa^{230} formed time to decay into U^{230} . At the end of this period, chemical separation of the uranium yielded essentially pure 20.8-day U^{230} into which its daughters soon grew. The sample of this material obtained was one previously prepared for measuring its alpha

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spectrum.³¹ The activity had been vacuum-sublimed onto a 0.002-inch platinum plate forming a band 1/8 inch by 1 inch. Since the principal measurements to be made were the alpha-gamma angular correlations, this sample was larger than desirable, but as in the case of Th²²⁸ other factors prevented observing the precise correlations, so that further efforts to reduce the sample size seemed unnecessary.

Previous work,³¹ using both chemical methods and recoil techniques to separate members of the decay family, identified the following gamma rays in the series:

U ²³⁰ (kev)	Th ²²⁶ (kev)	Ra ²²² (kev)	Ra ²²² (or daughters) (kev)
70	109	330	609
160	130	--	--
230	190	--	--
--	240	--	--

These assignments were confirmed by observing the gamma ray spectrum as a function of alpha particle energy in the following manner. A sodium iodide (thallium activated) crystal mounted in a vacuum chamber as has already been described was used to detect alpha particles and the spectrum so obtained for the U²³⁰ series is shown in Fig. 19. Each of these peaks is comprised almost entirely of the ground state transition which in some cases is considerably

Fig. 19. Alpha particle spectrum of U²³⁰ series
taken with a sodium iodide crystal.

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removed from the low abundance-low energy group which is coincident with the gamma ray of interest. Nevertheless, with a knowledge of the gamma ray under consideration, the alpha energy gate could be set at the proper place to minimize conflicting coincidences. For example, the 230-kev gamma ray of U^{230} decay and the 240-kev gamma ray of Th^{226} would not be resolved by the gamma ray spectrometer operating as a single-event instrument, but when the alpha particle gate was set at about 5.66 Mev (U^{230} maximum energy minus 230 kev) the 230-kev gamma ray showed a maximum coincidence rate and when the gate was set at about 6.10 Mev (Th^{226} maximum energy minus 240 kev) the 240-kev gamma ray showed up. Since the 240-kev peak is four times as intense as the 230-kev peak, there was still some overlap when the gate was set at 5.66 Mev. It was estimated that roughly 15 percent of the 230-kev peak was due to 240-kev gamma rays. In addition to confirming the assignments listed above, the 609-kev gamma ray was proved to belong to Em^{218} decay, which assignment was suspected but not determinable from the earlier measurements.

The proposed decay schemes for U^{230} and Th^{226} are shown in Fig. 20. These are based on alpha and gamma spectra and gamma-gamma coincidence measurements made previously³¹ and on the alpha-gamma angular correlations of the present study. It will be noted that the 230-kev state of U^{230} is shown as an unresolved doublet comprising the 1- and the 4+ states while in Th^{226} the 1- state is well separated from the others.

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Fig. 20. Decay schemes of U^{230} and Th^{226} .

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The angular correlations obtained by gating with the alpha particles of a particular energy and rotating the gamma ray detector, are shown in Figs. 21 and 22. In Fig. 21 the gate was set corresponding to 6.10 Mev which is the energy of $\text{Th}^{226} \alpha_{240}$ and the gamma ray peak at 240 kev shows a distribution with a minimum at 180° and maxima at 90° and 270° . This distribution is very similar to the one observed for the 217-kev gamma ray of Th^{228} decay, and by an argument identical with the one used in that case may be taken as good evidence for a 1^- assignment of the 240-kev level.

As seen in Fig. 21 a peak at 330 kev with a different correlation has appeared. This comes from coincidences with α_{330} of Ra^{222} (6.23 Mev) which, because of the poor resolution of the alpha peaks, overlaps considerably the gating energy (6.10 Mev). Because of this lack of discrimination and the fact that the 330-kev gamma ray is considerably more intense than the 240-kev one, the coincidence intensity of the 330-kev peak is likewise more intense. The angular correlation of the 330-kev gamma ray is consistent with that for a $0^+ \xrightarrow{\alpha} 2^+ \xrightarrow{\gamma} 0^+$ sequence as would be expected for an alpha transition to the first-excited state of an even-even nucleus.

The data in Fig. 22 were taken in the same manner except the alpha gate was set at 5.66 Mev which corresponds to α_{230} of U^{230} . A different gamma spectrum appeared and included was the 230-kev gamma ray showing the $0^+ \xrightarrow{\alpha} 1^- \xrightarrow{\gamma} 0^+$ correlation. The 160-kev peak involves the same energy level and its virtual absence of

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Fig. 21. Alpha-gamma angular correlations for the 240-kev gamma ray of Th^{226} decay and the 330-kev gamma ray of Ra^{226} decay.

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Fig. 22. Alpha-gamma angular correlations for the following gamma-ray transitions: 70, 160, and 230 kev from U^{230} decay; 330 kev from Ra^{222} decay.

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angular correlation is consistent with our supposition that the 230-kev level is a doublet of a $4+$ and a $1-$ state and therefore the sequence measured is a mixture of $0+ \rightarrow 1- \rightarrow 2+$ and $0+ \rightarrow 4+ \rightarrow 2+$. Other evidence that the 230-kev level is actually a pair of close-lying levels will not be reviewed here in detail. Briefly, part of the argument lies in the well-established systematic spacing of the $4+$ state which, for U^{230} decay, should be at about 230 kev. Also, the abundance ratio of the 160-kev gamma ray to the 230-kev gamma ray appears to differ by a factor of two between the alpha decay of U^{230} and the beta decay of $Ac^{226.57}$. If true, this can mean only that there are two levels populated to different degrees by the two modes of decay.

The abundant 70-kev transition of U^{230} decay also shows up in Fig. 22 with the $0+ \rightarrow 2+ \rightarrow 0+$ correlation expected, as does the 330-kev gamma ray of Ra^{222} decay appearing in low intensity.

B. Odd-Mass Nuclei

There are no systematics of odd mass number nuclei similar to those existing for the even-even type. The isotopes studied were therefore selected more or less at random, and will be presented roughly in order of decreasing mass number.

1. 99^{253} decay. -- 99^{253} is a 20-day alpha emitter with an alpha particle energy of 6.64 Mev.¹¹⁻¹⁵ It may be prepared by intensive neutron irradiation of lower elements as has been described in the

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section concerning 100^{254} . Two different samples were used in the present measurements. The first of these, which contained 7000 alpha disintegrations per minute of 99^{253} and about 200 beta disintegrations per minute of 36-hour 99^{254} , was mounted on a 0.0005-inch aluminum plate and was used for a determination of the L x-ray intensity. The second sample was milked from purified californium (Cf^{253} decays by emitting a negatron to 99^{253}) and was therefore of considerably greater isotopic purity. This sample contained about 6×10^4 alpha disintegrations per minute, and was vacuum-sublimed on to a 0.002-inch platinum plate. It was used both in the alpha particle spectrograph and for determination of the gamma ray spectrum.

A cursory examination showed that the only radiations of appreciable intensity associated with the alpha decay of 99^{253} were L x-rays. Their intensity was obtained by comparing the alpha-L x-ray coincidence rate with that of Am^{241} in which case the value of 0.41 L x-ray per alpha particle obtained by Beling et al.²⁰ was used. Corrections were made for differences in absorption of neptunium and berkelium L x-rays in the sample backing plate and detector window. The result indicated that 5.3 ± 1.0 percent of the 99^{253} alpha transitions were accompanied by L x-rays. In order to convert this number into primary electron vacancies, several assumptions must be made. First it was assumed that the yield of L x-rays per L vacancy is about 0.4. This will depend on the type of transition involved, and from the data of Kinsey²¹ is

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probably too low for an E2 transition, but about right for an M1. It must further be assumed that the ratio of L conversions to total conversions is about 0.8, which is found to be the case more or less independent of transition type in the heavy element region. Using these values it may be estimated that 17 percent of the alpha population is followed by a converted transition. This number may be compared with a rough value of 19 percent expected from the alpha particle data.

Figure 23 shows the gamma ray spectrum of 99^{253} observed on the sodium iodide (thallium activated) scintillation spectrometer. In order to reduce background radiation, the spectrum shown in Fig. 23a was taken in coincidence with alpha particles, which were detected at about 30 percent geometry with a zinc sulfide screen. Included here is the region from about 0 to 500 kev, and peaks are seen at 65, 112, and 393 kev. The radiation at 65 kev is very probably due to platinum K x-radiation from fluorescence excitation of the sample backing plate, and the 112-kev peak corresponds very closely in energy to what one would expect for berkelium K x-rays. The 393-kev gamma ray is of quite low intensity; however, in a long run on the weaker sample it was also discernible in about this same abundance. Since rigorous chemical separations were made on both samples, it seems very likely that this radiation is associated with 99^{253} . It might be mentioned that the grand-daughter, Cf^{249} , has a prominent gamma ray of almost this same energy. However, it also has a peak at 341 kev, and, since this is not observed in the

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Fig. 23. Gamma ray spectra of 99^{253} . a) High-energy region observed in coincidence with alpha particles. b) Low-energy region.

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present measurement, the 393-kev gamma ray cannot be ascribed to this isotope. The lower energy region of the gamma ray spectrum which is shown in Fig. 23b could not be observed in coincidence with alpha particles because of the absorption of the gamma rays in the 0.002-inch platinum plate. Therefore, the background radiations were large, and consequently the statistics were rather poor. The indicated limits in Fig. 23b represent the probable error. In addition to the L x-rays, peaks are seen at 43, about 56, 67, and approximately 110 kev. The 67- and 110-kev peaks are interpreted again as platinum and berkelium K x-rays. The 43-kev peak was also seen on an alpha-gamma coincidence run with the weaker sample and from this an intensity was obtained. The intensity of the 56-kev gamma ray was estimated by comparison with the 43-kev peak from the curve in Fig. 23b. The best values for the energies and intensities of the radiations are given in Table IV.

Table IV
Gamma Rays of 99^{253}

Energy	L x-ray	43 ± 2	56 ± 4	112 ± 3	393 ± 3
Intensity (%/α dis.)	5.3 ± 1.0	0.09 ± 0.03	0.02 ± 0.01	0.03 ± 0.01	0.04 ± 0.01

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The spectrograph runs⁵⁸ made on the second sample showed the alpha particle energy of the main group of 99^{253} to be at 6.642 ± 0.005 Mev. Two additional groups were seen 43 and 98 kev lower in energy with abundances of 9 and 5 percent, respectively. These values are only approximate due to the small amount of the activity available. Additional measurements of the alpha-particle spectrum have been made by Ghiorso⁵⁹ using an ion chamber, and he reports a group 390 kev lower in energy than the main group in an intensity of 0.05 to 0.1 percent.

The decay scheme of 99^{253} suggested by the alpha particle and gamma ray data is shown in Fig. 24. The low-energy gamma rays are in good agreement with the energy levels observed on the alpha particle spectrograph. Furthermore, by comparing the gamma ray intensities with the alpha populations to the levels, rough conversion coefficients of 150 and 250 may be calculated for the 43- and 56-kev transitions, respectively. Although these numbers are very poor, they seem to indicate predominantly M1 or E2 transitions, since if they were E1, the conversion coefficients would be about 100 fold lower, if M2, about 5 to 10 fold higher, and if the multipole order were E3 or higher, the gamma rays (or L x-rays from their conversion) would not have been found in coincidence with alpha particles (resolving time about 8×10^{-6} second). It would have been difficult to observe the 98-kev crossover transition due to the more abundant berkelium K x-rays, so that its absence may not be significant. This grouping of three close-lying energy levels

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Fig. 24. Decay scheme of 99^{253} .

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appears repeatedly throughout the odd mass number nuclei; a case very similar to the present one being U^{233}_{60} .

The 393-kev gamma ray fits well both as to energy and abundance with the low-energy alpha group observed subsequently by Ghiorso. If it is assumed that the K x-rays observed arise from the conversion of this gamma ray, then a K conversion coefficient of around 0.8 is indicated. This is sufficient to rule out electric transitions, but does not give a clear choice between M1 and M2.

2. Cf^{249} decay. -- Cf^{249} is an alpha emitter with a half-life of 400 to 500 years and an alpha particle energy of 5.80 Mev.^{22, 23} It may be prepared by intensive neutron irradiation of lower elements as has been discussed in previous sections. However, in order to reduce the amounts of $Cf^{250-252}$ present, the sample used here was milked from the separated berkelium fraction. Ultimately this should give isotopically pure Cf^{249} . Due, however, to the long half-life of Cf^{249} and a small amount of original californium present in the berkelium fraction, the sample available for these studies contained 33 percent Cf^{249} , the remainder being $Cf^{250-252}$. The sample was mounted on a 0.00025-inch platinum plate, and contained a total of about 3000 alpha disintegrations per minute.

The gamma ray spectrum observed in coincidence with alpha particles is shown in Fig. 25. Peaks are seen at about 60 kev, 105 kev (5 percent), 341 ± 4 kev (15 percent), and 394 ± 4 kev (62 percent), where the intensities given are with respect to Cf^{249} .

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Fig. 25. Alpha particle-gamma ray coincidence spectrum of Cf^{249} .

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alpha particles. The 60- and 105-kev peaks are probably platinum and curium K x-rays, respectively, whereas the other two must represent gamma ray transitions. The assignment to Cf^{249} seems certain since the gamma ray spectrum of $\text{Cf}^{250-252}$ mixtures has already been examined in this manner, and has shown nothing in intensities comparable with those observed here. The small essentially constant background is due to spontaneous fission-gamma ray coincidences from the $\text{Cf}^{250-252}$ present. It has previously been shown that these coincidences do not give rise to any peaks in this region.

In coincidence with L x-rays the two most prominent peaks observed were the K x-rays at 105 kev and the 341-kev gamma ray. Also seen in lower intensity was the 394-kev gamma ray and a previously unobserved peak at 265 kev. Although coincidences were run with the high-energy peaks in order to determine the energy of the transitions giving rise to the coincident L x-rays, these efforts were not successful, which probably indicates that the conversion coefficients are large.

Ghiorso⁶¹ has observed the alpha particle spectrum of Cf^{249} in an ion chamber equipped with Lektromesh and in addition to the main group at 5.80 Mev, groups higher in energy by 113 kev (approximately 6 percent) and 385 kev (about 3 percent) were found. This indicated that some 90 percent of the transitions lead to an excited state around 390 kev above the ground state. The present observation of high-intensity gamma rays of about this energy is consistent with this finding.

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An incomplete decay scheme for Cf^{249} is shown in Fig. 26. The 394-kev gamma ray almost undoubtedly corresponds to the transition between the most heavily populated level and the ground state. Although the 341-kev photon could originate from a level at 341 kev and drop to the ground state, the arrangement indicated in Fig. 26 is preferred on the basis that the 341-kev photon is more likely to compete with the one at 394 kev than would be one of some 50 kev. Direct alpha population to a level of 341 kev is ruled out by the intense 341-kev-L x-ray coincidences observed. From the number of K x-rays seen it may be concluded that both the high-energy gamma rays are electric in nature, since any magnetic transition would be more heavily K converted than is allowed. The state higher in energy than the one of 394 kev is inferred from the existence of low intensity 394-kev-L x-ray coincidences. There does not seem to be sufficient evidence to place in the decay scheme the 265-kev photon observed in coincidence with L x-rays. Also there is no definite indication as to the method of de-excitation of the 272-kev level.

3. Am^{243} decay. --A decay scheme recently proposed⁶² for Am^{243} is shown in Fig. 27. The spins of Am^{243}_{63} and Np^{239}_{64} are the result of spectroscopic measurements, and the alpha groups indicated were obtained on the alpha particle spectrograph.⁶² In addition, the 75-kev gamma ray has been shown to be present in 80 percent of the alpha transitions, which means a maximum conversion

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Fig. 26. Decay scheme of Cf^{249} .

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Fig. 27. Decay scheme of Am^{243} .

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coefficient of 0.25 and therefore an E1 transition.⁶⁵ This decay scheme is essentially identical with that of Am^{241} ⁶⁶ except for the following three points. First, the top three levels in Am^{241} decay are each moved down about 15 kev so that the prominent E1 transition is 60 instead of 75 kev. Secondly, although the measured spin of Am^{241} ⁶⁷ is 5/2 like that of Am^{243} , the ground state of Np^{237} ⁶⁸, unlike that of Np^{239} , is known to be 5/2. The final difference is in the lifetime of the state giving rise to the E1 transition, which is 6×10^{-8} second for Am^{241} ⁶⁹ and $< 2 \times 10^{-9}$ second for Am^{243} .⁷⁰

The alpha-60-kev gamma ray angular correlation in Am^{241} is known to be isotropic when measured under ordinary conditions. Fraser and Milton,⁷¹ however, have shown that this is due only to the long lifetime of the intermediate level, and using a coincidence circuit with a short resolving time, they find an initial correlation of $W(\theta) = 1 - 0.21 \cos^2 \theta$, which decays with a half-life of about 5.5×10^{-9} second. Novey,⁷² using liquid film sources, has obtained this same correlation. Now since, in the case of Am^{243} decay, the lifetime of the intermediate level is known to be less than 2×10^{-9} second, it seems quite reasonable that the alpha-75-kev gamma ray angular correlation should be subject to measurement using ordinary non-liquid samples and coincidence equipment of relatively long resolving time. The present work represents such a measurement.

The sample available was one obtained from the extensive neutron irradiation of Pu^{239} . The integrated neutron flux was such

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that the ratio of Am^{243} to Am^{241} by alpha activity was about 5 to 1. There was also some $\text{Cm}^{242-244}$ present in the sample, but this should not interfere with the measurements made. About 5×10^5 alpha disintegrations per minute of this material was vaporized onto a 0.006-inch aluminum plate over an area of $1/8 \times 1/8$ inch, and this constituted the principal source used. A second source was prepared by vaporizing about the same amount of activity onto thin mica. The reason for preparation of this source will be discussed later.

Since it was estimated that a considerable length of time would be required for each position of the angular correlation, a procedure was developed for taking data from four angles simultaneously. This amounted to having four potassium iodide crystal alpha particle detectors about $3/16$ inch in diameter mounted onto the ends of lucite light pipes 2.5 inches long by $3/8$ inch in diameter. Each of these light pipes was fastened by a grease seal to a Dumont-6292 photomultiplier tube, the output of which was fed through a preamplifier in the usual manner. At this point the pulses from the four preamplifiers were mixed and the total pulses amplified and fed into the signal side of the 50-channel pulse-height analyzer. Now, by controlling the high voltage on each of the phototubes individually, it was possible to distribute the alpha peaks among the 50 channels available such that there was about 10 to 12 channels per peak. The long lucite light pipe reduced the energy resolution to about 10 percent, but even so the peak-to-valley ratio between adjacent peaks was a factor of 50. The four potassium iodide alpha

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detectors were then placed around the Am^{243} source at fixed angles within 180° , so that alpha particles emitted in a particular direction would strike a given crystal. At another variable angle a gamma ray detector was employed, from which pulses corresponding to 75 kev were selected to gate the 50-channel pulse-height analyzer. By integrating the four alpha peaks observed, the number of coincidences in each of the four directions could be obtained. These were corrected for geometry by observing the alpha peaks directly in a separate run. Changing the angle of the gamma ray detector gave data on four different angles.

Four sets of data or a total of sixteen points were taken in this manner and the resulting angular correlation is shown in Fig. 28. The statistical probable error is shown on the extreme right hand point, and is comparable with that for all the others. Since it was not possible to count accurately the number of 75-kev gates due to interference from the Np^{239} daughter, the sets of points were normalized to one another. This could be accomplished by normalizing the sum of all coincidences observed in each set of peaks, the experimental arrangement having been such that the four points of each set would be symmetrically distributed on the correlation, whose rough shape was predetermined by the fact that a dipole radiation was involved.

The solid line shown in Fig. 28 corresponds to an angular correlation of $W(\theta) = 1 - 0.10 \cos^2 \theta$, which represents the least

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Fig. 28. Alpha particle-75-kev gamma ray
angular correlation in Am^{243} decay.

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squares fit of the data. However, corrections should be made for the finite solid angles subtended by the detectors which were 1/2 and 1 percent of the total sphere for the alpha and gamma sides, respectively. Rough calculations showed that these corrections would make the correlation $W(\theta) = 1 - 0.11 \cos^2 \theta$.

There are two other problems which need to be considered before the interpretation of the above result is discussed. These are the effect of the 11 or 12 percent of alpha population which goes to the 118- and 173-keV levels and subsequently cascades down to the 74-keV level, and the possibility that the correlation is being attenuated to some extent in a manner similar to that in Am^{241} . To get at the first of these problems, the angular correlation was rerun in exactly the same manner as previously except that the additional requirement was imposed that the alpha-75-keV gamma ray coincidence must also be in coincidence with an L x-ray in order to be registered. This enabled us to look directly at the interfering correlation, which after correction for the abundance of these events could be subtracted from the overall correlation of Fig. 28. Although the counting rates were quite low, leading to poor statistics, it was found that the data obtained in the above manner were isotropic to within 15 or 20 percent. This means that the angular correlation of alpha particles directly populating the 74-keV level becomes $W(\theta) = 1 - (0.126 \pm 0.03) \cos^2 \theta$.

The second of the problems mentioned above has to do with the possibility of the nucleus being reoriented between the alpha

and gamma ray emission so that the initial correlation is to some extent destroyed. Either a magnetic field operating on the magnetic moment of the intermediate nucleus or an electric field gradient on the electric quadrupole moment might have this effect. In an attempt to reduce this partial destruction should it exist the sample vaporized onto mica was prepared. It was mounted in a vacuum chamber and the alpha particles were counted through the mica. This presumably left the intermediate nucleus recoiling through vacuum, where it might be expected that appreciable electric and magnetic fields would not exist. Under such circumstances the points at 90° and 180° were checked, and within the statistical limits a result identical with that of Fig. 28 was obtained. Thus, as was earlier suggested, it would seem that the $<2 \times 10^{-9}$ second half-life of the intermediate state is sufficiently short to prevent any attenuation of the type found in Am^{241} decay.

If the 75-kev gamma ray terminates at the ground state (spin $1/2$) of Np^{239} , then since it is an E1 transition the only possible spin assignments for the 74-kev level are $1/2$ and $3/2$. The fact that any alpha-gamma angular correlation is observed immediately eliminates $1/2$ and fixes the spin of the intermediate level as $3/2$. Furthermore if the alpha particle is emitted with $l = 1$ and a change of parity, the theoretical correlation⁴⁶ for the $5/2-3/2-1/2$ sequence would be $W(\theta) = 1 - 0.14 \cos^2 \theta$, in excellent agreement with the experimental result. The assumption of any

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higher angular momentum value of the alpha particle will not give a correlation consistent with the experimental one (mixtures of angular momentum values have not been considered). This result implies that the ground states of Am^{243} and Np^{239} have the same parity.

There is, however, a bothersome point in the interpretation given above. The 74-, 118-, and 173-kev levels of Am^{243} have generally been considered to be members of a Bohr-Mottelson rotational band.⁶⁵ This is due to regularities in their level spacings and alpha populations. On such an assumption the spin of the lowest level may be calculated (from the level spacing) to be 2.5 ± 0.2 .⁶² This is inconsistent with the fact that an E1 transition proceeds to a level of spin 1/2 and with the value of 3/2 indicated from the angular correlation measurement. A possible solution of this difficulty suggested by Rasmussen⁷³ is that there is more than one level near the ground state, so that the 74-kev photon does not terminate at the spin 1/2 level. It would seem that either such a solution must be accepted or the Bohr-Mottelson interpretation of the three levels given up.

It is interesting to compare this case with Am^{241} , where again the level spacings give 5/2 for the spin of the 60-kev level. This time, however, the ground state spin is 5/2, and the angular correlation observed may be interpreted to be consistent with the expected 5/2-5/2-5/2 sequence either by assuming a p-wave alpha transition as in the present case, or by assuming a mixture of s-wave and d-wave transitions.⁷⁴

4. Am²³⁹ decay. --Due to the very marked similarity between the decay schemes of Am²⁴³ and Am²⁴¹, it was of considerable interest to examine the alpha decay of 12-hour Am²³⁹. Although the alpha branching ratio is only 0.003 percent, a sample was made available to us which contained roughly 200 alpha disintegrations per minute, and by means of alpha particle-gamma ray coincidences the radiations from the electron capture decay could be eliminated. There was a small amount of Am²⁴³ in the sample, and also an extremely small quantity of Am²⁴¹, but as will be seen these did not interfere seriously with the measurements.

In order to keep the chance coincidences small relative to the trues, only about 10 percent of the above sample was used in the coincidence run. The alpha particles were detected with a zinc sulfide screen at 35 to 40 percent geometry, and a beryllium window sodium iodide crystal served as the gamma ray detector. The spectrum obtained is shown in Fig. 29, where peaks of 17, 48, and 75 kev are seen. The 75-kev gamma ray is very probably that of Am²⁴³, which, as mentioned, was known to be present in the sample. The prominent peak at 48 kev is interpreted as a gamma ray in Am²³⁹ decay, and the 17-kev peak is undoubtedly a combination of neptunium L x-rays and the escape peak of the 48-kev photon. A subsequent coincidence run where the photons were detected with a xenon proportional counter established the energy of the 48-kev transition as 48.3 ± 1.5 kev. The large limit of error here is due to the fact that the statistics were quite poor. By integrating the 48-kev

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Fig. 29. Alpha particle-gamma ray coincidence spectrum of Am^{239} .

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peak of Fig. 29 and comparing it with the total alpha particles (corrections being made for gamma ray geometry and the Am^{243} alpha particles) a rough intensity of 0.5 ± 0.1 photon per alpha disintegration was found. This is similar to the values of 0.4^{20} and 0.8^{65} for the 60- and 75-kev gamma rays of Am^{241} and Am^{243} , respectively, and clearly identifies the transition as E1. By estimating the contribution of the escape peak to the 17-kev peak, and correcting for absorption in the backing plate and detector window, a very rough value of 0.3 L x-ray per alpha disintegration was obtained.

A comparison of the prominent features of the alpha decay schemes of the three americium isotopes is shown in Fig. 30. Assuming a reasonable conversion coefficient of around one for the 48.3-kev transition, it is seen that essentially 100 percent of the alpha transitions go through the 48-kev level as is the case for the analogous levels in the other two isotopes. The regular decrease in energy of the E1 transition with mass number is striking, and will be further discussed at a later time.

5. U^{235} decay. --An enriched sample containing 99.96 percent by mass of U^{235} was available for the present studies. Although only 0.022 percent by mass of U^{234} was in the sample, it was calculated that about 40 percent of the alpha activity was due to this isotope. This large alpha impurity was unfortunate as will be seen. It was, of course, not possible to obtain the alpha spectrum of U^{235} .

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Fig. 30. Comparison of the decay schemes of Am^{243} , Am^{241} , and Am^{239} .

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on the spectrograph, since the specific activity is too low. However, a number of gamma ray and coincidence measurements were made.

No chemical separations were required since the Th^{231} daughter in no way interfered with the experiments to be described, and no other impurity except uranium, itself, was present. The small samples used were mounted on 0.006-inch aluminum plates, and the large one was contained in a small glass-stoppered test tube.

The alpha-gamma coincidence spectrum is shown in Fig. 31. The coincidence technique was employed both to eliminate background radiation and to prevent the beta-emitting daughter, Th^{231} , from interfering. Peaks in this spectrum are observed at 185 ± 2 , 143 ± 2 , 109 ± 3 , and 93 ± 3 kev, with relative abundances of 100, 19, 6, and 9, respectively. The large amount of U^{234} alpha activity in the sample makes the absolute abundances considerably less reliable than these relative values given above. However, estimates that should be good to about 10 percent are 70, 13, 4, and 6.5 percent of the U^{235} alpha disintegrations, respectively. The 93-kev peak is, with little doubt, composed of thorium K x-rays. The two high-energy peaks represent previously recognized gamma rays of U^{235} , ⁷⁵ whereas the 109-kev photon has not been reported before. Probably this is because it is so close in energy to the K x-ray peak that slightly poorer energy resolution would have made it indistinguishable.

The large sample of U^{235} was used in a search for higher energy gamma rays of 289 and 386 kev reported to be associated with U^{235} .

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Fig. 31. Alpha particle-gamma ray coincidence spectrum of U^{235} .

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decay.⁷⁵ These were not found and a limit of 0.1 percent can be set on any gamma ray higher in energy than about 250 kev. Due to the existence of reasonably high-intensity coincidences that will be described below, it is probable that the reported peaks were due to simultaneous arrival in the detector of two photons.

The gamma ray spectrum in coincidence with the 185-kev photon, showed a gamma ray of 200 ± 10 kev (1.5 percent), K x-rays (5 percent) and L x-rays in an abundance greater than 13 percent. The L x-ray measurement was not carefully made, but the above minimum could be set. A correction was made in the intensity of the 200-kev peak for the low-energy shoulder caused by 185-kev photons which were gated by 200-kev gamma rays striking the gate detector. Coincidences with the 143-kev photon showed this same high-energy spectrum both with respect to energy and abundance. In addition, however, there was evidence for a peak around 40 kev and the L x-rays were considerably more intense than those found associated with the 185-kev peak. The absence of the 109-kev gamma ray from both these spectra was definite. Coincidences with the 109-kev photon and the K x-rays could not readily be interpreted, due to the large amount of daughter and Compton scattered radiation beneath these peaks.

Ghiorso,⁷⁵ using an ion chamber, has measured the alpha particle spectrum of U^{235} . He finds groups at 4.58 Mev (10 percent), 4.47 Mev (approximately 3 percent)?, 4.40 Mev (83 percent), and 4.20 Mev (4 percent). These groups together with the data presented above determine the decay scheme shown in Fig. 32.

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Fig. 32. Decay scheme of U^{235} .

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If it is assumed that 85 percent of the alpha disintegrations (the source of this figure will shortly become apparent) eventually pass through the 185-kev level, then absolute intensities of the 200-kev photon and K x-rays from the 385-kev level may be calculated to be 1.3 and 4.2 percent, respectively. The total, 5.5 percent, is in fair agreement with the alpha population of 4 percent to this level measured by Ghiorso. Furthermore, the indicated K conversion coefficient of around 3 gives the best agreement with an M1 transition, in which case the L conversion would be small and not aggravate the already large radiation intensity from the level.

Subtraction of the 4 percent K x-rays assigned in the preceding discussion from the 6 or 7 percent initially present, leaves only 2 or 3 percent to be attributed to the conversion of the 185- and 143-kev transitions. This makes an E1 assignment certain for the 185-kev transition and very probable for the 143-kev one, as well. The placement of the 143-kev photon as originating from the 185-kev level stems from the similarity of the spectra coincident with the two gamma rays, and from the fact that one would expect the 143-kev transition rather than one of some 40 kev to compete with the one of 185 kev. Including the 2 percent K x-rays, a total of about 85 percent of transitions from the 185-kev level can be accounted for. (The following suggested assignment of the 109-kev gamma ray as arising from this level may raise this number to around 89 percent.) This is in excellent agreement with Ghiorso's value of 87 percent of the alpha particles which should pass through

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this level.

The 109-kev gamma ray is shown in Fig. 32 as proceeding from the 185-kev level. This placement is by no means certain, but is arrived at by the following reasoning. It is likely that this transition is E1 in nature since even a conversion coefficient on the order of 3 (M1) would indicate around 16 percent abundance, which is difficult to place anywhere in the decay scheme. It is further likely that it lies around or below the 185-kev level, since it is not in coincidence with radiations from that level. The most reasonable arrangement, therefore, would seem to be the one in which the three E1 transitions compete from the 185-kev level. The indication of intense L x-ray coincidences with the 109-kev photon is consistent with this conclusion. A possible alternative would be a level at 109 kev, which would be in better agreement with the questionable alpha group assigned by Ghiorso⁷⁶ to a level of nearly this energy. However, this alpha group is very possibly due to an instrumental effect, and furthermore such an arrangement does not explain the L x-ray coincidences.

The level just higher in energy than 185 kev is implied by the intense 185-kev-L x-ray coincidences. Many of the high-intensity alpha transition in odd-mass nuclei of this region are found to populate members of a triplet structure ($\text{Am}^{243, 62}$ for example), and it seems quite possible that U^{235} does likewise. Since the gamma transitions within such a structure are highly converted the observation of only L x-rays in coincidence with the 185-kev photon would be explained.

6. Np²³⁷ decay. --About 1.5 mgm (2×10^6 alpha disintegrations per minute) of Np²³⁷ was available for the present study. This material had been prepared by an (n, 2n) reaction on U²³⁸, and was of high isotopic purity. The specific activity of Np²³⁷ is so low that examination on the alpha particle spectrograph was not feasible, so that the measurements made were concerned only with the gamma rays.

To remove the prolific radiations associated with the daughter, Pa²³³, a simple chemical separation was devised. The activity, in concentrated HCl, was placed on a column consisting of Dowex-A-1 anion resin. Under such circumstances both neptunium and protactinium are adsorbed onto the resin. Elution with 5 M HCl solutions, was then found to remove the neptunium rather quickly, while leaving the protactinium to come down quite slowly and even then only after a considerable period of time. The solutions containing Np²³⁷ were evaporated to dryness, redissolved in water, and transferred to 0.006-inch aluminum plates. Portions of up to 2×10^5 disintegrations per minute were purified in this manner.

Prominent in the gamma ray spectrum of these samples were L x-rays (43 percent) and peaks at 30 kev (12 percent) and 87 ± 3 kev (18 percent). The energy of the 30-kev radiation was measured in a xenon proportional counter and found to be 29.6 ± 0.5 kev. It is certain that the peak at 87 kev was composed principally of a gamma ray and not protactinium K x-rays, as later coincidence runs showed the K x-rays at 96 kev, distinctly shifted upward in

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energy from the peak observed here. Both of these low-energy transitions are, with little doubt, E1 in nature, simply by virtue of the fact that their abundance precludes sufficiently large conversion coefficients for any other multipole order. Two other peaks at 143 ± 3 and 198 ± 3 kev were seen in low intensity (0.5 to 1.0 percent), and were so broad as to indicate other even lower intensity radiations in the region. Nothing was observed above an energy of about 240 kev, however.

The gamma ray spectra in coincidence with the 30- and 87-kev photons were found to be identical except for the intensity of L x-radiation. About 40 percent of the 30-kev photons were found to be in coincidence with an L x-ray, while this number was only 17 percent for the 87-kev peak, a large part of which can be attributed to the K x-rays which are also present in this energy region. In both cases, however, K x-rays (18 percent), 143-kev photons (0.7 percent) and 193-kev photons (0.1 percent) were observed. The contribution of the K x-rays included in the 87-kev gate was expected to be small for these measurements. It was interesting that the amount of 193-kev radiation was much smaller relative to that of 143 kev than was observed in the gamma ray spectrum and furthermore the energy seemed lower than the previous 198 kev. This suggests that the two may not be the same gamma ray. The coincidence spectra were much cleaner also, so that no other radiations in the region were indicated.

The spectrum coincident with L x-rays was also measured. An 87-kev-K x-ray peak and the two at 143 and 193 kev were seen. Sufficient absorber was present to explain the absence of the 30-kev gamma ray. Once again other radiations were present in the higher energy region. In this case a single additional gamma ray of about 160-170 kev could account for the peak shapes, whereas in the gamma ray spectrum at least this radiation and one of some 230 kev were indicated.

There do not seem to be sufficient data available to construct a decay scheme, however several conclusions can be drawn. From the similarity of the spectra coincident with the 30- and 87-kev gamma rays it seems very likely that they originate from the same level. A large part of the L x-rays in coincidence with the 30-kev photon could well be from the conversion of the otherwise unobserved 57-kev transition. If so this is very similar to Am^{241} decay where El transitions of 60- and 26-kev proceed from a 60-kev level.⁶⁶ The analogy may be carried even further in that the 60-kev level in Np^{237} (from Am^{241} decay) has a lifetime of 6.3×10^{-8} second,⁶⁹ and conversion electrons in the presently considered spectrum have been found from a level with a half-life of 3.69×10^{-8} second.⁷⁸ Since, as was previously mentioned, the only prominent gamma rays in this spectrum are those of 87- and 30-kev, it seems very likely that again the level giving rise to the El transitions may be delayed.

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7. Th²³¹ decay. --Samples of Th²³¹ milked from highly purified U²³⁵ were used in a study of the radiations emitted by this nucleus. Although Freedman et al.⁷⁹ have proposed a decay scheme, certain aspects of this scheme seemed questionable and prompted the present investigation.

Chemical separation of thorium from uranium was achieved by stripping the thorium from a Dowex-A-1 anion resin column with 10 M HCl. Under such conditions uranium was found to remain adsorbed on the resin. The thorium was then transferred to 0.006-inch aluminum plates for gamma ray analysis.

L x-rays (100 percent) and gamma rays of 25.8 ± 0.5 kev (27 percent) and 85 ± 3 kev (30 percent) were the only prominent radiations observed. A xenon proportional counter was used to establish the energy of the 26-kev gamma ray. These radiations are strikingly similar to those of Np²³⁷ as, indeed, one might expect since the daughter nuclei are Pa²³¹ and Pa²³³. As in that case, the two gamma rays may again be immediately assigned as E1 transitions. Higher energy gamma rays were present in low abundance, but unfortunately sample size limitations prevented study of them.

Gamma-gamma coincidence work proved the bulk of the 26- and 85-kev photons were not in coincidence. This is contrary to the decay scheme of Freedman et al.,⁷⁹ however, by analogy to the Np²³⁷ case, was not totally unexpected. K x-rays in intensities of around 5 percent of the gates were found in coincidence with both

the 26- and 85-kev photons. No other high-intensity radiations were present (a search for L x-rays was not made). Again the low-intensity high-energy region could not be investigated.

Although a considerable amount of information is available on the beta components and gamma rays of Th^{231} , it is not possible to determine a unique decay scheme. It is tempting to again consider that the 85- and 26-kev photons arise from the same level, however there is as yet little evidence for this. Along this line it would be very interesting to look for a delay between beta particles and the gamma ray spectrum, similar to that found in Np^{237} and Am^{241} decay.

8. Th^{229} family decay. --Due to the limited amount of information which was available on the decay schemes of Th^{229} and the members of its family, and to the availability of a sample containing about 10^7 alpha disintegrations per minute of the parent, Th^{229} , a study of this group of nuclides was undertaken. The sample had been prepared by the neutron irradiation of Th^{232} to give Th^{233} , which after two negatron emissions yielded U^{233} . Initial chemical separation of the uranium should remove the target, Th^{232} , and subsequent milkings of thorium should give only the daughter, Th^{229} . However, it was found that the present sample contained in addition to Th^{229} , appreciable quantities of Th^{232} and Th^{228} , presumably from an incomplete initial separation of uranium from thorium. The interference caused by these impurities will be discussed individually for the members of the family.

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8a. Th²²⁹. --One of the primary objects of the investigation of Th²²⁹, itself, was observing its complex alpha spectrum on the alpha particle spectrograph. Unfortunately this was not possible because the mass of Th²³² present prevented preparation of a sufficiently thin sample. The only data obtained, therefore, were concerned with the gamma ray spectrum.

Chemical purification of Th²²⁹ from its decay products was achieved by extraction with benzene solutions of toluyltrifluoroacetone (TTA). Using a concentration of 0.4 M TTA it was found that with the aqueous phase adjusted to a pH of 0.9 to 1.0, the thorium alone extracted into the TTA-benzene layer. Subsequent removal of the TTA could be accomplished by back-extraction into 4 N HNO₃. This offered a fast and simple method of preparing samples for gamma analysis.

The gamma ray spectrum contained three peaks at 87, 148, and 200 kev, whose relative intensities were very roughly 100, 5, and 15, respectively. L x-rays were also observed, but due to absorption uncertainties an intensity was not obtained. The 87-kev peak is probably composed chiefly of radium K x-rays, however coincidence studies of this peak showed it to be in coincidence with a peak of similar energy to an extent of about 10 percent, so it is possible that a gamma ray of nearly this same energy exists. Other radiations in coincidence with this peak were in low abundance, however the indications were that the 148-kev gamma ray was present and the 200-kev one was not. On the other hand coincidences with the L x-rays showed in addition to a large peak at 87 kev, indications of coincidences with

both the 200- and 148-kev photons. Little interference with these studies was expected from the low-abundance radiations of Th^{228} , itself, and its daughters were removed by the chemical separation. It might be mentioned that gamma ray spectra taken several days after purification showed the prominent 238-kev photon of Pb^{212} (from Th^{228}).

Complexity of the alpha spectrum of Th^{229} was found by Hagemann et al.,⁸⁰ who observed groups at 5.02 Mev (10 percent), 4.94 (20 percent), and 4.85 (70 percent) using an ion chamber. However, with the meagre amount of data available on this isotope, it does not seem to be possible to propose a decay scheme.

8b. Ra^{225} . --The immediate daughter of Th^{229} is the 14.8-day beta emitter, Ra^{225} . Chemical purification of radium could be achieved again by extraction with 0.4 M TTA solutions in benzene. Using a pH of about 5.0 it was found that radium alone remained in the water layer, all other activities present having been extracted into the TTA-benzene phase. Still present, however, from the Th^{228} in the original sample was Ra^{224} . To remove it, the radium fraction was allowed to stand for about a month, during which time the 3.64-day Ra^{224} essentially decayed out. Repurification then yielded pure Ra^{225} .

The gamma ray spectrum from a sample so prepared is shown in Fig. 33. Of the two peaks seen the larger, at 40 kev, is interpreted as a gamma ray of Ra^{225} , while the other, at 14 kev, is probably

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Fig. 33. Gamma ray spectrum of Ra^{225} .

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composed of actinium L x-rays and the escape peak of the 40-kev radiation. Subsequent analysis on a xenon proportional counter established the energy of the gamma ray to be 40.0 ± 1.0 kev. Coincidences with beta particles of energy about 50 to 300 kev showed the same gamma ray to be present, and from this, its intensity per beta disintegration was determined to be 29 percent. This allows a total conversion coefficient no larger than 3.5, and at once clearly indicates an E1 transition. After correction for absorption and for the escape peak of the 40-kev photon, an L x-ray intensity of 13 percent per disintegration was found. If an overall L-shell fluorescence yield of 0.5 is assumed, and if the reasonable assumption that the L x-rays arise from the conversion of the 40-kev photon is made, then an L conversion coefficient of 0.9 may be calculated for the gamma ray. This constitutes even stronger evidence for the E1 nature of the 40-kev transition. Using a value of 0.78 for the ratio of L conversion to total conversion (this is the number found for the 60-kev E1 transition in $\text{Am}^{241, 66}$) the total conversion coefficient becomes 1.1, and the population to the 40-kev level may be calculated to be 63 percent. There is no evidence for population of any higher levels, as no higher energy gamma rays were seen (limit 0.5 percent), and no radiations at all were found in coincidence with the 40-kev photon.

Both the singles beta spectrum and that in coincidence with the 40-kev photon were taken on an anthracene scintillation spectrometer. In each case an endpoint of 0.32 ± 0.03 Mev was obtained.

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Due to difficulties in determining accurate beta endpoints, this method was not expected to show any difference between those of the singles and coincident spectra. The purpose of the measurements was a rough determination of the beta energy which agrees well with the value of 0.31 Mev obtained by Freedman et al.⁸¹ These authors also observed the conversion lines of the 40-keV photon.

A final measurement on Ra^{225} was of the half-life of the 40-keV level. By analogy to the 60-keV level in Np^{237} ,⁶⁹ the 27-keV level in Ac^{227} ,⁸² and a level of uncertain energy in Pa^{233} ,⁷⁸ all of which are de-excited by E1 transitions and have measurable lifetimes, it seemed reasonable that the level in Ac^{225} from which the 40-keV E1 transition proceeded should similarly have a measurable lifetime. This, however, was found not to be the case, and a limit of $<2 \times 10^{-9}$ second was set for the half-life of this level.

The decay scheme of Ra^{225} indicated by these data is shown in Fig. 34. The log ft values of 7.2 and 6.9 for the ground and 40-keV states, respectively, may be calculated from the known half-life, the rough beta energy, and the abundance of population to the two levels. By the Nordheim selection rules⁸³ both of these transitions are first forbidden ($\Delta I = 0, 1$; with a parity change). This is not possible, since the levels are known to have different parity. A similar difficulty is found in Pu^{243} decay,⁸⁴ where about equal beta population to levels connected by an 85-keV E1 transition is observed. These difficulties along with others (Np^{238} , for example--to be discussed later) indicate that extreme caution should

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Fig. 34. Decay scheme of Ra^{225} .

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be used in applying simple beta selection rules to obtain spins in this region.

8c. Ac^{225} . --The next member of the family was the 10-day alpha emitter Ac^{225} . In this case the daughter, Fr^{221} , grew in with only a 4.8-minute half-life so that chemical separation from decay product activities was not feasible. Furthermore, the beta emitter, Ra^{225} , did not interfere either with the alpha particle spectrograph measurements or with the gamma ray studies, which were done by alpha-gamma coincidence techniques, so no radium-actinium separation was made. It was, however, necessary to remove thorium from the sample in order to be able to prepare a thin spectrograph source. This was done by the same method that was used to purify the thorium, itself, namely TTA extraction at a pH of 0.9 to 1.0. Most of the thorium-free material was then vacuum-sublimed onto a 0.002-inch platinum plate for analysis in the alpha particle spectrograph. A small portion was vaporized in a similar manner onto 0.006-inch aluminum for gamma ray studies to be described presently.

The alpha particle spectrum is seen in Fig. 35. The main group, present in 54 percent abundance, was found to have an energy of 5.815 ± 0.010 Mev by comparison with Po^{218} , whose alpha particle energy Briggs³³ has determined to be 5.998 Mev. In addition, lower energy groups which lead to excited states of 37, 100, and 190 kev were observed with abundances of 29, 13.5, and 3.5 percent,

Fig. 35. Alpha particle spectrum of Ac²²⁵.

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respectively. The relatively poor energy resolution was due to sample thickness which in turn was due to massive amounts of material (not thorium) left from the chemical separations.

Simple alpha-gamma coincidences would not give the gamma ray spectrum of Ac^{225} due to the presence of alpha-emitting daughters. It was therefore necessary to use a potassium iodide crystal as the alpha detector, so that the alpha particles of the various emitters could be separated by their different energies. Then by gating only with those alpha particles due to Ac^{225} , the gamma ray spectrum shown in Fig. 36 was obtained. In order to avoid platinum absorption difficulties, the sample vaporized onto aluminum was used. Peaks are seen at 60, 85, 150, 187, and 215 kev, and in addition there is a distinct shoulder on the high-energy side of the 85-kev peak which can only be interpreted as a gamma ray of about 100 kev. Assignment of the 215-kev peak to Ac^{225} must be considered improbable due to the high abundance 220-kev gamma ray of the daughter, Fr^{221} . The francium alpha group giving rise to this radiation is only 300 kev removed from the actinium alpha groups so that with an alpha energy resolution of 6 percent sufficient overlap to give the small peak seen in Fig. 36 is considered quite reasonable. Also it was concluded that the 60-kev group did not belong to Ac^{225} . Its intensity was found to vary as a function of time and gating energy compared with those of the other peaks seen. Furthermore, a relatively small amount of Am^{241} could have caused this peak, and the alpha energy of this isotope is sufficiently close to Ac^{225} that it would not have

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Fig. 36. Pulse-height analyzed alpha particle-gamma ray coincidence spectrum of Ac^{225} .

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been eliminated in this manner. The other four peaks can be assigned to Ac^{225} with reasonable certainty, the one at 85 kev being francium K x-rays and the other three gamma rays.

The problem of gamma ray intensities is a difficult one in this case. They cannot be obtained from Fig. 36 and the number of alpha gates recorded because all the Ac^{225} alpha particles were probably not counted. In a total alpha-gamma coincidence run using a zinc sulfide screen to detect alpha particles there is interference in the K x-ray region from daughter activities present. Also in this type of experiment the 150-kev gamma ray is so small compared with the one at 220 kev from Fr^{221} , that it can barely be observed. However, by this method estimates can be made which indicate that the K x-rays of francium are present to an extent of 3 to 4 percent per Ac^{225} alpha disintegration, the 100-kev gamma ray to about 1 to 2 percent, and the 150- and 187-kev radiations to roughly 0.8 and 0.5 percent, respectively. Although it is again emphasized that these are only estimates, it will be seen that they are fairly consistent with the alpha populations in the decay scheme to be proposed.

Figure 37 shows a decay scheme consistent with the data described. The 187- and 150-kev gamma rays fit well as transitions from the level populated by the lowest energy alpha group to the ground and 37-kev states, respectively. The energy of 187 kev obtained from the gamma ray data is probably somewhat better than the 190 kev indicated by the alpha particle spectrum for this level. Since the 100-kev radiation is very likely below the K edge of francium, it seems probable that all the K x-rays come from

Fig. 37. Decay scheme of Ac^{225} .

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conversion of the two higher energy gamma rays. This accounts for a total of around 5 percent population to the 187-kev level, which agrees well within the estimated limits of error with the 3.5 percent measured on the alpha particle spectrograph. The large number of K x-rays compared to photons observed for these transitions indicates that at least one and probably both are magnetic in nature.

The 100-kev gamma ray very likely represents the transition from the 100-kev level to the ground state. From the 13.5 percent alpha population to this level, a conversion coefficient of 5 to 10 might be estimated for transition. This is not at all inconsistent with an E2 or M1 transition, but due to the approximate nature of the gamma ray intensities and the distinct possibility of unobserved radiation to the 37-kev level it cannot be considered very meaningful. It probably should be mentioned here that some 87-kev radiation corresponding to the transition from the 187- to the 100-kev levels could easily be present. This would affect the above arguments, in particular those related to the K x-ray abundances. However, due to the lack of evidence for such a transition, the best assumption seems to be that it does not occur in appreciable intensity.

No radiation of about 37 kev was seen, so that the de-excitation of the 37-kev level was not directly observed. However, if the transition were anything besides E1 in nature, one would not expect to observe the photons because of the large conversion coefficient.

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For this reason a search for L x-rays was made to see if their intensity was large enough to account for about 30 percent L and lower electron vacancies from this source, plus another 10 to 15 percent from the higher energy transitions. This measurement is even more difficult than the gamma ray intensities, since absorption losses, fluorescence yields, and other uncertainties in addition to those already discussed are encountered. It can, however, be stated that results indicating 30 to 50 percent L and lower electron vacancies were obtained. This number is sufficiently large so that no discrepancy with the decay scheme of Fig. 37 is indicated.

8d. Fr²²¹. --Using the sample whose preparation has already been described, the alpha particle spectrum of Fr²²¹ was measured. Two groups were found whose energies were 6.332 ± 0.010 Mev (83.8 percent) and 6.116 ± 0.010 Mev (16.2 percent). These data are in reasonable agreement with the earlier ion chamber work of Hagemann et al.⁸⁰ Adding in the difference in recoil energy, a level in At²¹⁷ of 220 ± 14 kev is indicated.

The gamma ray spectrum was observed by two different techniques. Total alpha-gamma coincidences gave the combined spectra of all the alpha-emitting daughters in the Ra²²⁵ family. However, no gamma ray was present in sufficient intensity to interfere with an abundance determination of the prominent 220-kev photon of Fr²²¹.⁸⁵ By this method an intensity of 12.7 percent of Fr²²¹ alpha

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disintegrations was obtained. By pulse-height analyzing the alpha particles, and selecting only those of Fr^{221} as gates, a clean gamma ray spectrum was observed. From this an energy of 220 ± 2 kev was measured for the photon, and a ratio of K x-rays to 220-kev photons of 0.23 was calculated. This presumably gives an upper limit of the K conversion coefficient of the gamma ray, since as has been mentioned the alpha energy resolution is such that small amounts of the more abundant K x-rays from Ac^{225} might be included. The total conversion coefficient may be calculated from the alpha population to the level and the photon abundance to be 0.28. Table V compares the experimental conversion coefficients with those expected of E1, E2, and M1 transitions. The theoretical values are from the screened calculations of Rose et al.⁸⁶ M and lower conversion coefficients are not available, but are not expected to be large. The transition is seen to give the best agreement with an E2 assignment.

Table V
220-Kev Gamma Ray of Fr^{221}

	E1	E2	M1	Experimental
a_K	0.06	0.15	1.53	<0.23
$a_{K+L} \approx a_{\text{total}}$	0.07	0.35	1.81	0.28

8e. At²¹⁷ and Po²¹³. --The alpha particles of At²¹⁷ and Po²¹³ were also observed and the energies measured to be 7.051 ± 0.010 Mev and 8.351 ± 0.010 Mev, respectively. These are in fair agreement with the previous ion chamber measurement,⁸⁷ but seem consistently about 15 to 20 kev higher in energy. No complexity of either alpha spectrum was found, and likewise no gamma rays encountered which could be assigned to either isotope. In the latter case, however, no intensive search was made, so that a low limit cannot be set.

8f. Bi²¹³. --Bi²¹³ was known to be a beta emitter (2 percent alpha branching) with a half-life of 47 minutes. Wagner et al.⁸⁵ found two components in the beta spectrum of 1.39 Mev (68 percent) and 0.959 Mev (32 percent). Consistent with this was their finding of a 0.434-Mev gamma ray. Only one measurement can be added to these data. The number of K x-rays associated with Bi²¹³ was obtained and from this a K conversion coefficient of 0.2 was calculated. This is in good agreement with that expected for an M1 transition.

8g. Tl²⁰⁹. --Gamma rays associated with the alpha decay of Bi²¹³ could not be observed, however those of the daughter of this process, Tl²⁰⁹, were present in reasonably high abundance. Wagner et al.⁸⁵ found the beta energy of 2.2-minute Tl²⁰⁹ to be 1.99 Mev, and associated with this decay they found a gamma ray of 120 kev.

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By gating with beta particles of energy greater than 1.4 Mev, thereby eliminating all other members of the Ra^{225} family, a gamma ray spectrum of Tl^{209} could be observed. Such a measurement showed peaks at 0.08, 0.12, 0.45, and 1.56 Mev. The radiations around 0.08 Mev are very probably K x-rays of lead, whereas the other three must represent gamma rays. By turning this process around and gating with the photons, it was possible to prove that each of these gamma rays was indeed associated with the 1.99-Mev beta spectrum of Tl^{209} . Figure 38 shows the beta spectrum in coincidence with the 1.56-Mev photon together with that in coincidence with the 0.43-Mev photon of Bi^{213} for comparison purposes. The low abundance, high energy tail on the spectrum in coincidence with the bismuth gamma ray extends out to about 2 Mev and is due to the 0.45-Mev radiation from Tl^{209} which overlaps considerably the gating energy.

Gamma-gamma coincidence studies indicated that each of the three gamma rays was in coincidence with the other two. For example, the spectrum coincident with the 0.12-Mev photon is shown in Fig. 39. Peaks at 0.45 and 1.56 Mev are readily observable. These data show that the three gamma ray transitions are in cascade, so that together with the beta energy of 1.99 Mev, a decay energy of 4.12 Mev is calculated. This is in reasonable agreement with the value obtained from closed cycles which is 3.92 Mev.⁸⁸

The position of the transitions in the cascade may be gotten from work by Harvey,⁸⁹ who investigated the energy levels of Pb^{209} from

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Fig. 38. Beta spectra in coincidence with the
1.59- and 0.43-Mev gamma rays
of Tl^{209} and Bi^{213} , respectively.

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Fig. 39. Gamma ray spectrum coincident with the 0.12-Mev photon of Tl^{209} .

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(d, p) reactions on Pb^{208} . He found levels at 1.56 and 2.03 Mev (among others) which are in excellent agreement with those obtained by placing the two highest energy gamma rays observed here lowest in the cascade. The 0.12-Mev transition must therefore be on top, indicating a level at about 2.13 Mev, which was not observed by Harvey. The decay scheme is shown in Fig. 40.

Since the ratio of K x-rays to 0.12-Mev photon was observed to be the same whether gated with high-energy beta particles or with either of the other two gamma rays, it was concluded that the major source of the K x-rays was very probably the conversion of the 0.12-Mev gamma ray. This gives a K conversion coefficient of 0.4, which if the above assumption is wrong, represents a maximum value. A search was made for L x-rays, and none were observed, the limit set being such that the L conversion coefficient is less than one. This leads to the rather surprising result that the most likely assignment for the 0.12-Mev transition is E_1 .

C. Odd-Odd Nuclei

In each case the odd-odd nuclei studied here decayed by beta emission to nuclei of the even-even type. This process is of considerable interest in that it frequently gives information on excited states higher in energy than those populated to an appreciable extent by the alpha emission of other even-even nuclei. The most extensive data are available on Np^{238} , which, for this reason, is discussed first.

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Fig. 40. Decay scheme of Tl^{209} .

1. Np²³⁸ decay. --There is a considerable amount of information available on the decay scheme of the 2.1-day beta emitter, Np²³⁸.^{90, 91} The present coincidence studies represent a part of a rather thorough investigation of this isotope,⁹⁰ which has led to the decay scheme that will be shown somewhat later. First a review of the previous data pertinent to this study is in order.

Beta spectroscopic work^{90, 91} has shown two beta components, 1.27 Mev (45 percent) and 0.27 Mev (55 percent), along with six gamma rays of energy 44.0, 102.2, 986, 1029, 942, and 927 kev. The last two of these are considerably weaker than the preceding two, whose relative abundances have been shown to be $\gamma_{986}/\gamma_{1029}$ equals 0.83/1.00 from uranium photoconversion electron measurements.⁹⁰ The intensity of conversion electrons from the 44- and 102-kev transitions were found to be approximately 70 and 3 percent of the beta disintegrations, respectively,^{90, 91} and due to known large conversion coefficients,¹⁸ these numbers represent roughly the intensities of the transitions. Cm²⁴²¹⁸ decays by alpha-particle emission to the same nucleus, Pu²³⁸, from which it was known that low-lying levels at 44- and 146 kev exist. The 102- and 44-kev transitions are also seen in this decay and correspond to the transitions from the 146-kev state to the 44-kev state and from the 44-kev state to the ground state, respectively. It is from this source that the conversion coefficients of these two low-energy transitions are known to be large.

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A sample of Np^{238} which had been prepared by neutron irradiation of Np^{237} was available for the present studies. The integrated neutron flux was such that no significant amount of Np^{239} was formed. Portions of this sample varying in size from 10^4 to 6×10^5 beta disintegrations per minute were evaporated from solution onto 0.006-inch aluminum plates for the present study.

The first point to be cleared up by the coincidence work was the amount of branching of the high-energy beta particles to the ground and 44-kev states. This could be done by measuring the beta-L x-ray coincidence rate and comparing it with alpha-L x-ray rate in Cm^{242} , which decays to the same nucleus with a known 26.3 percent branching to the 44-kev level.¹⁸ Regions of the beta spectrum away from conversion electrons and from the low-energy component (endpoint, 0.27 Mev) were required as gates, and for this purpose the intervals 0.41 to 0.61 Mev and 1.02 to 1.20 Mev were selected. Some difficulties were encountered when high activity levels were used, however, with samples of sufficiently low activity a limiting value of 100 ± 5 percent to the 44-kev level was obtained from both intervals. Because the measurement was made by comparison with Cm^{242} mounted in a similar manner it is independent of such uncertain quantities as fluorescence yield, absorption losses, and geometry.

A similar measurement was made gating with the high-energy gamma rays. In this case it was necessary to correct for the 3 per-

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source rather than from direct beta population). The results then indicated that of the approximately 55 percent hard gamma rays, 3 percent go to the 146-kev level (assumed), 32 percent to the 44-kev level, and 20 percent to the ground state.

These two results indicate at once that 80 ± 5 percent of the beta transitions pass through the 44-kev level. This is to be compared with values of 65⁹⁰ and 75⁹¹ percent for the intensity of the conversion electrons from the 44-kev transition. The disagreement is not considered to be serious due to counter window transmission and sample thickness difficulties in the beta spectroscopic work.

The question now arose of which of the high-energy gamma rays were populating particular low-energy states. To get at this, detailed comparisons were made of the singles gamma ray spectrum in the region of 1 Mev with those in coincidence with 100-kev photons and with L x-rays. There are K x-rays included in the 100-kev peak, but these will not contribute coincidences with high-energy gamma rays, therefore this coincidence spectrum should show the hard gamma rays populating the 146-kev level. The L x-rays arise predominantly from conversion of the 44-kev transition, and thus coincidences with them will enhance the high-energy photons which populate the 44-kev level. Figure 41 shows the result of this comparison. Figure 41a is the spectrum in coincidence with the 100-kev photons, and clearly either the 927- or the 942-kev gamma rays or both are seen. Figure 41b includes both the singles spectrum and that in coincidence with L x-rays. The noteworthy observation is

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Fig. 41. Gamma ray spectra of Np^{238} .

a) Coincident with 100-kev radiation.

b) — singles; --- coincident with
L x-rays.

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that the highest energy portion of the peak is depressed when observed in coincidence with L x-rays. This is taken to indicate that part or all of the 1029-kev gamma ray proceeds to the ground state. Independent 100-kev photon-gamma ray coincidences by Rasmussen⁹⁰ confirm the first part of these results.

Finally, it was desired to determine the source of the population of the 146-kev level. It was certain that some came from the high-energy gamma rays as shown in the preceding discussion, but what amount of the total 3 percent population this comprised was unknown. There are two ways to obtain this information. The first is by looking for a hard beta component in coincidence with the 102-kev gamma ray, and the other is by quantitative measurement of the high-energy gamma ray-102-kev photon coincidences. Both of these methods were tried, neither, however, very successfully. Figure 42 shows the beta spectrum in coincidence with radiation of about 100 kev. The conversion electron peak at approximately 900 kev is the result of K x-rays included in the gating energy. Since a conversion coefficient of 5 has been measured for the 102-kev transition,¹⁸ only about 0.5 percent photon per beta disintegration is expected. It happens that the K conversion lines of the high-energy gamma rays are also present in 0.5 percent abundance,⁹⁰ so that if all of the population to the 146-kev level were by beta particles, the integrated hard beta spectrum should be just equal to the integrated conversion electron peak in Fig. 42. This does not seem to be the case; although unfortunately the tailing between the conversion

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Fig. 42. Beta spectrum of Np^{238} in coincidence with 100-kev photons.

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electron line and the soft beta spectrum is so large that a significantly low limit on beta population to the level cannot be set.

The other method of determining the mode of population to the 146-kev level is by measuring the number of 100-kev photons in coincidence with the high-energy gamma rays. If a sufficient intensity is obtained to indicate that all of the 0.5 percent arises from this source direct beta population is excluded. Lead K x-rays from fluorescence excitation of absorbers used interfered with this measurement, so that the problem was not resolved. An opportunity of repeating this experiment with non-lead absorbers has not yet been available.

Two assumptions are necessary before a unique decay scheme can be constructed. Since there are no data on the relative abundance of the 927- and 942-kev gamma rays, it must be assumed that they are present to the same extent as are their conversion electrons, namely $\gamma_{942}/\gamma_{927} \approx 2/1$.⁹⁰ Also, as was mentioned, it is uncertain whether one or both of these gamma rays populate the 146-kev level. It will be assumed that both do, which fixes the sum of their intensity as 3 percent of the beta disintegrations. This is in agreement with the amount of these photons expected from an attempted resolution of the gamma ray peak of Fig. 41b into its four components.

Consideration of the data together with these assumptions defines the level scheme shown in Fig. 43. The 1029-kev gamma ray is calculated to be present in 28 percent of beta disintegrations, only 20 percent of which goes to the ground state (as discussed earlier).

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BETA DECAY SCHEME OF Np^{238}

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Fig. 43. Decay scheme of Np^{238} .

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Therefore, a second gamma ray of very nearly this same energy is proposed, and it fits well from the level at 1073 kev to the 44-kev level. The intensities of the weaker transitions are quite uncertain and for this reason are enclosed in parentheses. The level at 1087 kev is considered uncertain since it is based only on the assumption that the 942-kev gamma ray terminates at the 146-kev level. In the paper by Rasmussen et al.,⁹⁰ an additional decay scheme is considered, which assumes that this gamma ray populates instead the 44-kev level.

The conversion coefficients of the two prominent high-energy gamma rays may be calculated from the intensities given in Fig. 43 and the conversion electron intensities measured on the beta spectrometers.^{90, 91} Values of 0.007 and 0.008 are obtained indicating that both are probably E2 transitions. This defines the spin and parity of the 1029-kev level as 2+, since the ground state is assumed to be 0+. The composite nature of the 1029-kev gamma ray is not expected to affect this argument seriously.

Consideration of the nucleus in terms of the Bohr-Mottelson^{9, 10} rotational model leads to the introduction of the K quantum number, which represents the projection of the total angular momentum, I, along the nuclear symmetry axis. For a state of I = 2, this can have values 0, 1, or 2. Calculation of the reduced transition probabilities of the gamma ray transitions from the 1029-kev level to the three lowest levels gives excellent agreement with those expected if the upper level has K = 2, and excludes the other two

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possibilities. For this reason the levels at 1029 and 1073 kev are considered to be members of a rotational band with $K = 2$ and spins, I , of 2 and 3, respectively. The detailed arguments relating to this assignment, and that of the parent, Np^{238} , are to be found in the paper by Rasmussen et al.⁹⁰

2. 99^{254} decay. -- 99^{254} is the parent of 100^{254} , and as such its preparation has already been discussed.¹¹⁻¹⁶ Quite recently an alpha-emitting isomer of 99^{254} with a half-life on the order of a few years has been discovered,¹⁶ however the present measurements are concerned with the 36-hour beta emitter. The beta endpoint had previously been found to be 1.1 ± 0.1 Mev.⁹²

The sample available for the present study was one previously described in connection with 100^{254} decay. It contained 7000 alpha disintegrations per minute of 99^{253} , and about 300 beta disintegrations per minute of 99^{254} in equilibrium with its daughter 100^{254} . Inasmuch as the two alpha emitters present have only low-energy, low-abundance radiations associated with their decay, they gave little interference with the present experiments. The sample was mounted on 0.0005-inch aluminum foil.

The gamma ray spectrum in coincidence with beta particles whose energy was between roughly 50 and 1500 kev was first examined. Figure 44 shows the high-energy portion of this spectrum in addition to which a region that included L x-rays (20 kev) was taken. Due to the possibility of impurities a half-life measurement

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Fig. 44. Gamma ray spectrum of 99^{254} observed in coincidence with beta particles.

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was made on all the observed radiations. Also from the number of gates recorded and the approximate geometry, intensities were estimated. However, the fact that background radiations were comparable with those from the sample makes these estimates particularly uncertain. The data are summarized in Table VI.

Table VI
Gamma Rays at 99^{254}

Energy (Mev)	Intensity (%)	$T_{1/2}$ (hours)
0.020 (L x-ray)	50	41
0.123	10	38
0.66	40	37

The total beta spectrum decayed with a 44-hour half-life indicating a possible impurity which may also have contributed a small number of beta-L x-ray coincidences. Although an attempt was made to discriminate against the soft conversion electrons of 99^{253} , it may be that a sufficient number of them was counted to give the longer half-lives noted.

In order to be certain that the 0.66-Mev radiation belongs with 99^{254} decay, the beta spectrum in coincidence with it was observed. One would expect this endpoint to be the difference between 1.1 and 0.66, or about 0.44 Mev. The observed endpoint was 0.45 ± 0.1 Mev.

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Both the total beta spectrum and the L x-ray-beta coincidences showed endpoints around 1.1 Mev. It thus seems almost certain that the 0.66-Mev radiation is associated with the decay of 99^{254} .

The gamma ray spectrum in coincidence with the 0.66-Mev photon was also examined. Only L x-radiation was observed, and it was present to an extent of about 50 percent per photon. A limit of 5 percent could be set as the maximum amount of radiation in the 100-kev region per gate photon. This limit will be of use in the following interpretation.

In order to construct a decay scheme from the small amount of data available, it is necessary to assume low-lying states of 100^{254} similar to those of other even-even nuclei in this region. Inasmuch as every such nucleus examined has shown the familiar $0+$, $2+$, $4+$... pattern,⁵ this assumption does not seem unwarranted. Levels are thus expected at about 44 and 145 kev together with cascading gamma rays of about 100 and 44 kev, whose conversion coefficients should be of order of 5 and 1000, respectively.

The decay scheme shown in Fig. 45 may be deduced from the data and the above assumptions. Using an overall L-shell fluorescence yield of 0.5, it is found that there is about one L vacancy in coincidence with each 0.66-Mev photon, suggesting that this gamma ray proceeds predominantly to the 44-kev level. The limit of less than 5 percent 100-kev photons per 0.66-Mev gate further shows that the 145-kev level is not heavily populated by this means. The hard beta component is shown populating the 44-kev state, although

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Fig. 45. Decay scheme of 99^{254} .

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there are other possibilities. The reason it is preferred this way is the fact that there is about 50 percent L x-ray (100 percent L vacancy) per beta particle. As there is about 100 percent L vacancy per 0.66-Mev radiation (and therefore presumably per soft beta particle) there still remains essentially one L vacancy per hard beta particle. This number is not good enough to eliminate moderate branchings to the 145-kev or ground states (or a combination of the two which could yield one L vacancy per beta particle), but seems to indicate that the 44-kev level is the one most likely populated.

There are at least two possibilities for the 0.123-Mev radiation. It could represent the transition between the 145- and 44-kev states, but is slightly higher in energy than is expected for this gamma ray. It is, however, almost exactly the energy expected for K x-radiation of element 100. Furthermore, its intensity is roughly what one might expect from the conversion of the 0.66-Mev gamma ray if this radiation is of the M1 or M2 type. This assignment is therefore preferred.

The decay scheme proposed for 99^{254} is basically quite similar to that suggested in Fig. 43 for Np^{238} . Many of the details brought out in the Np^{238} case by the intensive beta ray spectroscopy could be present also in 99^{254} decay but would not yet have been detected.

3. Eu^{154} decay. -- The only isotope examined that was not in the translead region was Eu^{154} . This was done primarily for two reasons. First, the rare-earth region is, in many respects, similar to the region of the very heavy elements, so that it was of interest to look for possible analogs of the 1- states found in even-even nuclei of the radium-

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thorium region. And secondly, a sample of purified Eu¹⁵⁴⁻¹⁵⁵ was readily available for these studies. This sample had been recovered from the fission fragments accumulated in the neutron irradiation of the heaviest elements.⁹³ The Eu¹⁵⁵ present contained no high energy radiations^{94, 95} and therefore did not interfere with the measurements made. About 10^6 beta disintegrations per minute of Eu¹⁵⁴ was available in solution, from which smaller samples were prepared as needed by evaporation onto 0.002-inch platinum plates.

The high-energy gamma ray spectrum is shown in Fig. 46. Gamma rays are identified at 0.25, 0.60, 0.73, 0.88, 1.01, 1.28, and 1.6 Mev. In addition a look at the low-energy region revealed gadolinium K x-rays, and a prominent gamma ray at 0.12 Mev. Another peak at 0.09 Mev was present, but this was believed due to Eu¹⁵⁵.⁹⁴ Grover⁹³ had previously examined the spectrum of this sample and found these same gamma rays and no others. Their assignment to Eu¹⁵⁴ was substantiated by the fact that they were all found to be interrelated by the coincidence studies.

Extensive gamma-gamma coincidence measurements were made on the above gamma rays. These were complicated by the fact that a Compton smear from the higher energy radiations will be present beneath each peak. This made many of the results difficult to interpret. However, in Table VII the non-ambiguous results are listed.

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Fig. 46. High-energy gamma ray spectrum of Eu^{154} .

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Table VII
Gamma-Gamma Coincidences in Eu^{154}

Gating photon	In coincidence	Not in coincidence
0.12 Mev	all gamma rays	--
0.25	0.76	--
0.73	0.88, 1.01	1.28, 1.6
1.01	0.60, 0.73	0.88, 1.01, 1.28, 1.6
1.28	0.12	all others

For the photons not listed in each case, it may be assumed that either the appropriate region of the spectrum was not examined or the results were not readily interpretable. Several comments on these results are in order. As indicated, all the gamma rays were found to be in coincidence with the 0.12-Mev photon, and, in fact, the relative intensities were not appreciably changed from those of Fig. 46 with the exception of the 1.01-Mev gamma ray. In this case the coincidence peak was only about half as large as the singles peak. Since the 0.12-Mev photon will later be shown to be from the first excited state to the ground state, the above result means that there must be two gamma rays of nearly 1.01 Mev, one of which goes to the ground state, and the other of which goes to the first excited state along with all the other radiations observed.

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Also it is of interest to point out that the 0.76-Mev radiation in coincidence with the 0.25-Mev photon showed a definite shift in energy from the usual 0.73-Mev peak seen. It is therefore believed to be a separate gamma ray whose intensity in the total gamma spectrum is rather low. Special efforts were made in this case to avoid spurious coincidences caused by Compton scattering.

Before going further it would be well to point out the following sums, which should be useful in the construction of a decay scheme.

$$\begin{array}{lll} 1.01 + 0.60 = 1.61 & 0.25 + 0.76 = 1.01 & 1.61 + 0.12 = 1.73 \\ 0.88 + 0.73 = 1.61 & 0 + 1.01 = 1.01 & 0.73 + 1.01 = 1.74 \\ 0 + 1.6 \approx 1.61 \end{array}$$

Each of the sums is expected to be significant as the appropriate coincidences are noted in Table VII.

Beta-gamma coincidences were not extremely useful due to the many beta components present. It was found, however, that the hardest beta particles present were in coincidence with the 0.12-Mev gamma ray in sufficient intensity to rule out appreciable direct beta population of the ground state.

From neutron capture work, Church⁹⁶ has found gamma rays of 0.1234 and 0.2477 Mev associated with Gd¹⁵⁴. These very likely correspond to the gamma rays of 0.12 and 0.25 noted here, and are taken to indicate levels in Gd¹⁵⁴ at 0.123 and 0.371 Mev, which, from their spacing, are assumed to be the 2+ and 4+ rotational levels. In addition, beta work done by several groups^{95, 99} shows components of the beta spectrum with endpoints at 0.25, 0.59, 0.90, and

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1.88 Mev. Since it has been shown that the hardest beta particles are in coincidence with the 0.12-Mev photon, the beta groups indicate levels at 0.12, 1.10, 1.41, and 1.75 Mev.

These data, together with the sums noted previously, readily suggest that decay scheme shown in Fig. 47. The solid lines indicate the observed beta components, and the broken lines are those which may be present in lower intensity. The observed component to a level at 1.10 Mev probably represents a mixture going mostly to the 1.14-Mev level with some contribution to the 1.01-Mev level. The fit of the present gamma ray and coincidence data with the previous beta ray work is quite satisfying. It should be added that several of the higher energy photons discussed here had previously been identified.^{97, 98}

The similarity of the lowest five levels of the decay scheme in Fig. 47 with those of Np^{238} (Fig. 43) is indeed striking. In addition, from the gamma ray spectrum coincident with the 0.73-Mev photon the relative intensities of the 1.01- and 0.88-Mev gamma rays from the 1.01-Mev level may be obtained. This intensity is very close to what one would expect if the 1.01-Mev level had $K = 2$, $I = 2$ like the 1.029-Mev level of Np^{238} . On this basis the spin assignments shown were made, and it should be emphasized that they are tentative. No state was found which could be given a 1-assignment.

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Fig. 47. Decay scheme of Eu^{154} .

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IV. DISCUSSION OF RESULTS

A. Even-Even Nuclei

1. Rotational states. --It is well established that throughout the entire periodic table almost all the first excited states of even-even nuclei have spin 2 and even parity.¹⁻⁵ In the region well above the closed shells of 82 protons and 126 neutrons additional low-lying states are observed which together with the ground (0+) and 2+ states are generally considered to be members of a rotational band according to the Bohr-Mottelson unified collective and individual particle nuclear model.^{4, 9, 10} Of interest here is the fact that the levels of such a band are expected to have spins 0+, 2+, 4+ ..., and lie at energies proportional to $I(I + 1)$ where I is the spin of the particular level under consideration. In addition, the overall spacing of the band is expected to become progressively wider as the region of the closed shell is approached.

The extensive data of Asaro and Perlman^{4, 99} on the even-even alpha emitters conform remarkably well to the above situation. In every case where the measurements could be made the levels corresponding to the 0+, 2+, and 4+ states were found. Beyond this point the intensity of alpha population to the levels is so low that the results are very difficult to obtain; however, in several cases states corresponding to the energy expected if the spin were 6+, have been detected. Furthermore, the energy of the 2+ state, which reflects the overall spacing of the band, has been found to be constant at about 40 to 45 kev in the transuranium region, below

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which it rises rather sharply to a maximum value of around 2.5 Mev at the closed shell (lead) and then decreases to another minimum in the rare-earth region.^{3, 4} Experimentally the spins of the levels above the 2+ state are uncertain; however, they are generally considered to be 4+ and 6+ due to the agreement of their energy with that calculated from the $I(I + 1)$ relationship and to the fact that no evidence to the contrary exists (crossover transitions within a band, for example). Recent angular correlation work in the case of Th^{230} ⁵⁴ has given added support to the 4+ assignment in that case.

With this in mind the results of the heavy even-even nuclei studied here may be considered. In each case a level around 44 kev was observed which decayed to the ground state by a transition which was shown either to be, or to be consistent with, an E2 assignment. Thus a 2+ level is implied which agrees not only in spin assignment but in energy as well with those in the immediate preceding region.

Similarly 100-kev radiation was found in all three cases (100^{254} , $\text{Cf}^{250-252}$ and Cf^{256}), which was taken as evidence of a level around 145 kev in excellent agreement with curium and plutonium isotopes.⁴ The ratio of 145 to 44 (3.3) is exactly that calculated from the $I(I + 1)$ relationship for a 4+ and 2+ state, respectively.

Additional regularities established by Asaro and Perlman^{4, 99} have to do with the alpha populations to the various members of the rotational band. They have shown that if reasonable values for the nuclear radius are assumed, the populations to the ground state, and with somewhat less precision to the 2+ state, are just what would

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be calculated from simple one-body alpha-decay theory (coulombic barrier penetration). On the other hand the 4+ and 6+ states receive considerably less population than would be calculated on that basis. This situation is generally described as hindered decay, and a hindrance factor may be calculated which represents the factor by which the observed population is lower than that calculated. Figure 48 is a plot of the known hindrance factors to these rotational states against atomic number. The points for the decay of californium and element 100 are taken from the presently described work, and the others, for the most part, from the work of Asaro and Perlman.⁹⁹ There is no immediately apparent reason either why the hindrance to a given level should vary in the manner observed or why the variation among the levels should be so large. However, Rasmussen¹⁰⁰ in considering the quadrupole interaction of outgoing alpha particle waves has reached the conclusion that neither of these effects is particularly unexpected.

2. 1- states. --The low-lying 1- states found in even-even nuclei of the radium-thorium region were of considerable interest in that analogous levels do not seem to be found either above or below this region. Five cases have been identified with relative certainty and these are listed in Table VIII together with the gamma rays from the levels. In addition it was suggested that a level in Em^{222} (from Ra^{226} decay) of 255 kev might also belong to this group.

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Fig. 48. Hindrance factors to the rotational states of even-even nuclei. (This plot was prepared by F. Asaro.)

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A plot of the energy of the 1- states against neutron number is shown in Fig. 49. Here the points of a given Z have been connected. Although there is no a priori reason for plotting in this manner, a minimum is observed at 136 neutrons which may be of some significance.

A comment is here in order concerning the absence of the 1- levels from the alpha decay of higher elements (Cm²⁴², ¹⁸ for example). This could stem from either of two effects. From the plot of Fig. 49 it seems that the energy of this level might be quite high in regions removed from 136 neutrons. Under such conditions the maximum allowed alpha population (from one-body theory) to the level might be very low, so that its detection would be difficult. On the other hand even if the level were not at particularly high energies the hindrance factor might be sufficiently large to again reduce the intensity below detection. In this connection it should be added that of the 1- states observed, the hindrance factors are generally comparable with those of the 4+ states.^{4, 99} This statement, however, implies that they are rising with increasing mass number in this region as, indeed, they are. It would be of particular interest to examine the decay of some odd-odd beta emitters in this region since the limitations as to population of the level might not be so severe in these cases. One such examination, ⁵⁷Ac²²⁶ decaying by negatron emission to Th²²⁶, showed essentially complete decay to the 1- level.

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Fig. 49. Energy of the 1- states in even-even nuclei.

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In all five cases, E1 gamma ray transitions were found from the 1- state to both the 0+ (ground) and the 2+ states. The abundances of these radiations could be obtained from the sodium iodide scintillation spectrometer measurements, and by removing the expected third power Weisskopf energy dependence, reduced transition probabilities were calculated. Table VIII gives the result of this treatment.

Table VIII
Reduced Gamma Ray Transition Probabilities from 1- States

Parent alpha emitter	Daughter nucleus	Energy γ_0 γ_2		Reduced transition probabilities γ_0/γ_2
U^{232}	Th^{228}	324	270	0.45
U^{230}	Th^{226}	232	159	0.51
Th^{230}	Ra^{226}	253	184	0.49
Th^{228}	Ra^{224}	217	137	0.30
Th^{226}	Ra^{222}	242	130	0.48

γ_x represents the transition from the 1- state to the state of spin x.

The consistency of the data around a value of 0.5 for the ratio of the ground state to 2+ state reduced transition probabilities is unmistakable. Probably, however, the data are not so good as the

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figures would indicate. The value for Th^{228} seems distinctly low so that a review of the data was made. This indicated that about 20 percent error was not at all unreasonable, and there is no reason why the other values should not be subject to errors at least that large.

Nonetheless this should offer a basis for calculating the expected intensity of the unobserved approximately 67-kev transition to the 188-kev level in Ra^{226} decay. A value on the order of 4×10^{-4} percent is obtained for this intensity, to be compared with the experimental limit set of about 7×10^{-3} percent. Thus the statement which was previously made, that a significantly low limit could not be set, may be understood.

Finally, it is interesting to speculate as to the nature of such a level. Its existence is not readily understood from simple considerations. The ground state rotational structure would not be expected to give rise to states of odd spin or parity under normal circumstances, and a level based on the unpairing of two nucleons would scarcely be expected to occur at such a low energy. However, let us consider again the Bohr-Mottelson quantum number, K , which represents the projection of the spin, I , on the nuclear symmetry axis. For a level of spin 1, K may have values of either 0, or 1, and from the reduced transition probabilities of Table VIII a choice between these two possibilities may be made. If K is 1, the ratio γ_0/γ_2 would be expected to be 2.0, whereas if K is 0, this ratio becomes 0.5. The agreement of the data with the value 0.5 has

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already been noted, so that the assignment, $K = 0$, seems clearly indicated. The meaning of this result is not entirely clear at present, however, since K for the ground state rotational band is 0, a connection between the two seems somewhat more likely. Bohr¹⁰¹ and Christy¹⁰² have commented that such odd-spin, odd-parity states might arise from the rotational band and be displaced upward in energy if the nucleus were pear-shaped. It should be quite interesting to investigate this possibility in view of the present result.

B. Odd-Mass Nuclei

One of the most unusual and interesting features among the odd-mass nuclei of the translead region is the impressive number of low-energy $E1$ transitions. This is particularly true for the odd proton nuclei, as is shown in Tables IX and X, where the available data have been collected.

Also shown in these tables is the half-life of the level giving rise to the $E1$ transition wherever such measurements have been made. Some of the levels, in Np^{239} and Ac^{225} , for example, have half-lives too short to be measured as would be expected for electric dipole transitions of this energy. On the other hand, those levels in Np^{237} , Pa^{233} , and Ac^{227} , have measurable half-lives which are over 10^5 longer than predicted by the Weisskopf formula.

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Table IX

El Transitions in Odd Z, Odd A Nuclei

Parent	Daughter	Parent level of the El transition			Reference
		Energy (kev)	Energy (kev)	Half-life (second)	
Pu ²⁴³	Am ²⁴³	85	85	--	84
Am ²⁴³	Np ²³⁹	75	75	$<2 \times 10^9$	65, 70
Am ²⁴¹	Np ²³⁷	59.6	59.6	6×10^{-8}	66, 69, 103
		26.4	59.6	6×10^{-8}	
Am ²³⁹	Np ²³⁵	48.3	48.3	--	--
Np ²³⁷	Pa ²³³	87	(87)	(3.7×10^{-8})	78
		29.6	(87)	(3.7×10^{-8})	
Th ²³¹	Pa ²³¹	85	--	--	79
		25.8	--	--	
Pa ²³¹	Ac ²²⁷	27	27	3.7×10^{-8}	82, 104, 105
Ra ²²⁵	Ac ²²⁵	40.0	40	$<2 \times 10^{-9}$	--

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Table X
El Transitions in Even Z, Odd A Nuclei

Parent	Daughter	Energy (kev)	Parent level of the El transition		Reference
			Energy (kev)	Half-life (second)	
Cm ²⁴³	Pu ²³⁹	104	381	--	77, 106
U ²³⁵	Th ²³¹	185	185	--	--
		143	185	--	--
		(109)	(185)	--	--
Th ²²⁷	Ra ²²³	50	--	--	107*
Tl ²⁰⁹	Pb ²⁰⁹	(120)	2030	--	--

*The El character in this case is inferred from the intensity of 0.15 photon per alpha disintegration given by this reference.

In order to explain the existence of these transitions it is necessary to have close-lying levels of comparable spin and different parity. Limiting the discussion for the present to the odd proton case, this would seem at first glance to be an unlikely situation, since in the entire 82 to 126 proton shell, the shell model allows only the $i_{13/2}$ orbital of positive parity, and no negative parity orbitals of spin larger than 9/2. A solution to this problem has been suggested by Rasmussen.¹⁰⁸ He has shown that in the region where the Bohr-Mottelson strong-coupling configuration predominates, the nuclear spin can be considerably lower than the

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j value of the single nucleon. This would allow the parity of the $i_{13/2}$ orbital to be brought in, without at the same time necessitating such a large spin. Further, it can perhaps account for the very slow E1 transitions as being due to the large change of orbital angular momentum involved.

Another feature of the decay schemes of odd-mass nuclei in this region has to do with the existence of apparent rotational bands, as has been discussed by Rasmussen,¹⁰⁸ Asaro and Perlman,^{65, 99} and Moore.¹⁰⁹ In these cases three levels are observed which conform most particularly in their level spacing to the expectations of a rotational band according to the Bohr-Mottelson^{9, 10} nuclear model. Specifically, levels are predicted with spins $I_0, I_0 + 1, I_0 + 2 \dots$, which are spaced according to the $I(I + 1)$ rotational energy dependence. From the three experimental levels, it is possible to calculate I_0 , and for the bands in Np²³⁷,¹⁰⁸ Np²³⁹,⁶⁵ and Th²²⁹,⁹⁹ a value of 5/2 is indicated in each case. Further regularities having to do with the gamma ray transitions within the band and the alpha population to its members have been noted in these cases and shown to be in accord with the theoretical expectations.^{110, 111} To the above isotopes it seems likely that Bk²⁴⁹ (99²⁵³ decay) should be added. Its decay scheme (Fig. 24) is quite similar to that of U²³³, and as such fulfills the conditions outlined above. Pu²³⁹, from Cm²⁴³ decay, was formerly thought to be a fifth member of this group, but recent data of Asaro and Perlman,¹⁰⁶ indicate that this now seems less likely.

Several other possible groups of levels have been considered in this regard,¹⁰⁹ for some of which I_0 is other than $5/2$; however, at present the evidence for these cases seems less convincing than for the ones mentioned above. It should also be recalled that in the case of Np^{239} (Am^{243} decay) the spin of $5/2$ obtained for I_0 by these arguments was in definite disagreement with some of the experimental data, so that caution should be used in assigning spins by this method.

A final point of interest is concerned with trends in the energy levels observed. In this respect the odd proton nuclei are much simpler than those with odd neutrons so that for the present, at least, the discussion will be limited to this type. It was pointed out in connection with Fig. 30 that the neptunium isotopes from the alpha decay of the americiums present a very uniform picture in their level structure. Both Np^{237} and Np^{239} have well-defined rotational bands, the lowest member of which decays by an $E1$ transition whose energy is about 15 kev smaller for Np^{237} . The small amount of evidence on Np^{235} confirms the trend of the $E1$ transition energy with mass number. This situation suggests that a plot of the energy levels against mass number might be useful for extrapolation to other neptunium isotopes.

Figure 50 is a plot of the type suggested above, except that it has been expanded to include all of the odd proton isotopes in this region. The assumption that such a simple relationship exists for nuclei of different proton number is somewhat surprising, and probably is not completely valid, as will be discussed presently; however, the fact

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Fig. 50. Tentative energy level trends in odd-proton nuclei.

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that the levels of Fig. 50 do seem to correlate is at least some justification for such an assumption. The data come only from reasonably well confirmed decay schemes (or parts thereof), for which references may be found either in Table IX or in earlier sections on the particular isotope. The relative parities within a single level scheme are, for the most part, well known, but between level schemes they are not known and were selected to give the most reasonable agreement with Fig. 50. In several cases states whose energies fall higher than the maximum energy of the plot (300 kev) are known; however, no levels within the energy range shown have been intentionally omitted.

Several interesting comments may be made in spite of the forthcoming criticism of such a simple plot. Most notable, perhaps, is the almost linear continuation in the actinium isotopes (225 and 227) of the level which was the ground state in the previously discussed neptunium region. Also in Am^{243} (Pu^{243} decay) this trend is maintained although the linearity no longer is. The absence of this level (indicated by the absence of any E1 transition) in Bk^{249} could conceivably be tied in with a proposed subshell at 96 protons.¹¹² Pertinent here is the fact that the $i_{13/2}$ orbital could be the one just filled at this point and removal of a particle thereafter (to obtain different parity states) might occur only at considerably higher energies. Berkelium, of course, has 97 protons and would lie beyond this subshell. Also of interest is the observation that the levels of negative relative parity shown in Fig. 50 fall into discrete

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groups whose energies are around 40, 100, and 200 kev. This is perhaps in part explained by the rotational bands which predict some of these spacings, but even in the lower mass number region, where rotational spacings should become wider, as for the even-even isotopes, these same energies seem to persist. Furthermore, a 200-kev level is not expected from such considerations.

Although it is felt that a plot of the type shown in Fig. 50 is very likely applicable for each element individually, the interconnection of levels from different elements by plotting against mass number is probably oversimplified. Evidence that this is the case may be obtained from the protactinium isotopes. Although the level structures of Pa^{231} and Pa^{233} are as yet uncertain, sufficient data are available to make the schemes predicted from Fig. 50 seem unlikely. Thus as more data become available, the type of plot which will prove most useful will very likely have to be considered further. It should be added here that the odd neutron nuclei seem in many respects to be subject to this same type of treatment. Many additional complications are present, however, and as consistent a picture as given in Fig. 50 cannot be constructed.

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