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ALPHA DECAY STUDIES IN THE FAMILIES  
OF THE LIGHT URANIUM ISOTOPES

Carl Phillip Ruiz  
(Thesis)

April 1961

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ALPHA DECAY STUDIES IN THE FAMILIES  
OF THE LIGHT URANIUM ISOTOPES

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ALPHA DECAY STUDIES IN THE FAMILIES  
OF THE LIGHT URANIUM ISOTOPES

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April 1961

ABSTRACT

Using a  $180^\circ$  double-focusing alpha-particle spectrograph, an alpha-particle ionization chamber, and scintillation spectrometers, the alpha and gamma radiations of the  $U^{228,229}$  series,  $U^{233}$ ,  $Pa^{229}$ , and  $Ra^{222}$  were investigated. Decay schemes are presented in most cases and analysed wherever possible in terms of the theoretical nuclear models. For the odd mass nuclides the Bohr-Mottelson model was used to interpret the decay schemes for  $A \geq 225$ ; for  $A < 225$  there was no obvious rotational pattern present. The spectroscopic data obtained for the even-even nuclides are in agreement with the previously existing systematics, and are discussed in terms of the axially asymmetric theory of Davydov, et al.

The half-lives of  $U^{228}$ ,  $Ra^{220}$ ,  $Rn^{216,217,218}$  were measured. Conventional counting techniques were used for the  $U^{228}$  half-life determination while delayed coincidence counting and moving tape techniques were employed for the others. The data for the even-even nuclides are in good agreement with the systematics.

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I. INTRODUCTION

The understanding of nuclear structure has been greatly increased during the past few years. In the system of nuclei, it is well known that in the region of closed shells nuclei are spherically symmetric while in others a spheroidal deformation is the equilibrium shape. The model of Mayer<sup>1</sup> and Jensen<sup>2</sup> provides a good description of the spherical nuclei, while the axially symmetric (but non-spherical) model of Bohr and Mottelson<sup>3</sup> has had considerable success in this other region. For even-even nuclei, the recent asymmetric rotor theory of Davydov et al.<sup>4</sup> shows promise in correlating the collective states occurring in the regions of transition between these two as well as in the spheroidally deformed regions. However, little is known about particle states in the transition region, either from experimental information or from theory.

Large contributions to the understanding of nuclear structure have been possible through the copious amount of data obtained in nuclear spectroscopic investigations. It is in the regions of transition where the nucleus changes from spherically symmetric to spheroidally deformed that the amount of data is seriously deficient. The present investigation was directed toward obtaining information on even-even and odd mass nuclides in the transition region above lead in order to further our understanding of this region. For this purpose the alpha spectroscopic study of the U<sup>228</sup> and U<sup>229</sup> families and U<sup>233</sup> was undertaken. The decay of Pa<sup>229</sup> and Ra<sup>222</sup> was also investigated since these nuclides were readily available from the production of U<sup>229</sup>.

## II. EXPERIMENTAL METHODS

In an experimental investigation there are often practical limitations in obtaining the information desired. For example, in problems involving nuclear spectra it is not always possible to use an instrument of high resolving power (low transmission) because the isotope under investigation cannot be prepared in sufficient quantity. In this research these considerations were often limiting.

In the case of the  $U^{229}$  family a barely adequate quantity of activity could be produced for study on the high resolution electromagnetic alpha particle spectrograph. For the  $U^{228}$  family an alpha particle ionization chamber had to be employed. The gamma spectra were studied with a scintillation spectrometer incorporated into a coincidence circuit. Details of these instruments as well as those used for measuring half-lives, and the methods used in reproducing and purifying the activities are given in the ensuing sections.

### A. Alpha Spectroscopy

#### 1. Alpha-particle spectrograph

The alpha spectra of  $U^{233}$  and the  $U^{229}$  family were obtained with the alpha particle spectrograph. This instrument was built following the calculations of Judd.<sup>7</sup> It is essentially a Siegbahn-Svartholm double-focusing spectrograph with the modification that the source and receiver are located outside the magnetic field. The magnet covers  $180^\circ$  with a maximum radius of 50 cm and an optical axis of 35 cm. The non-uniform magnetic field has a radial dependence which produces focusing in both vertical and horizontal directions. The equation for the magnetic field is given by

$$H = H_0 \left( 1 - \frac{1}{2\rho} + 0.2089 \rho^2 + 0.473 \rho^3 \right) \quad (1)$$

where

$$\rho = \frac{r - 35.0}{35.0}$$

and  $r$  = radial position in cm.

The magnet current power supply is constant to within one part in  $10^4$ . To achieve this, the control amplifier is surrounded by a water jacketed container which minimizes changes in temperatures to

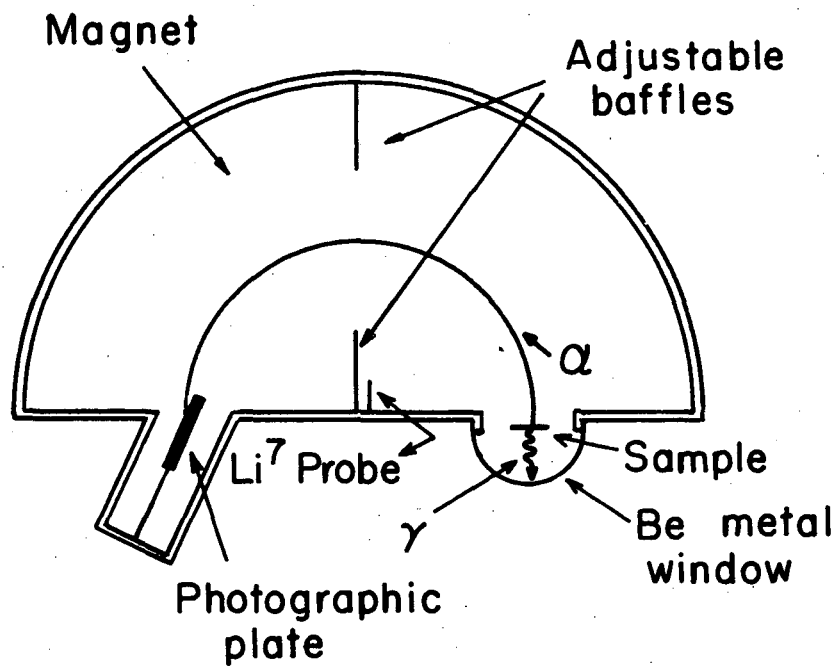
which the amplifier is sensitive. The current is monitored with a recorder and the magnetic field is measured in terms of the frequency of the nuclear magnetic resonance of  $\text{Li}^7$ . The spectrograph is maintained at a vacuum of  $10^{-5}$  mm of mercury during operation.

Figure 1 shows a schematic representation of the spectrograph. The alpha emitting source can be collimated by placing a defining slit directly over the plate containing the activity. The slit sizes vary from .002 inch to .25 inch depending on the nature of the experiment. The alpha beam is further defined by a pair of adjustable baffles located along the  $90^\circ$  radius. The transmission varies linearly with the baffle opening and reaches its maximum value of  $5 \times 10^{-4}$  of  $4\pi$  at a baffle opening of  $\pm 7$  cm.

After passing through the magnetic field, the focused beam impinges on a photographic plate at an angle of  $65^\circ$  to the normal of the beam direction leaving tracks about 20 microns in length. The plates, Eastman NTA, 25 micron, are  $1 \frac{15}{64}$  by 9 inches. Since the angle of acceptance in the vertical direction allowed by the gap between the pole pieces is small (about 1 inch), the alpha tracks on the plate should be nearly parallel.

After exposure, the developed emulsion is scanned with 450 power microscope with bright-field illumination. The plate is scanned along the short dimension and only those tracks which are parallel and of the proper length are accepted. The count is recorded as a function of position in the long dimension. Any tracks which do not meet the above specifications are rejected. This allows for partial discrimination between tracks which come from the source and those originating from contamination in the machine due to recoils emitted from the source. Since one microscope field of view is  $1/4$  by  $1/4$  mm, a complete counting of the plate requires a scan at every  $1/4$  mm position along the length of the plate. The transmission for particles which register across the long dimension of the plate has been shown to be constant.<sup>8</sup>

There is always a residual and fairly constant background of tracks on the plate even in regions quite distant from intense peaks. These tracks originate from contamination inside the machine (due to alpha radioactive recoils emitted by the source) as well as scattering of some of the alpha particles emitted at the source position. It was found that this background could be reduced by at least a factor of 5 by putting in another collimator having an opening  $1 \frac{1}{2}$  inches wide and placed about 4 inches



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Fig. 1. Schematic diagram of alpha particle spectrograph.

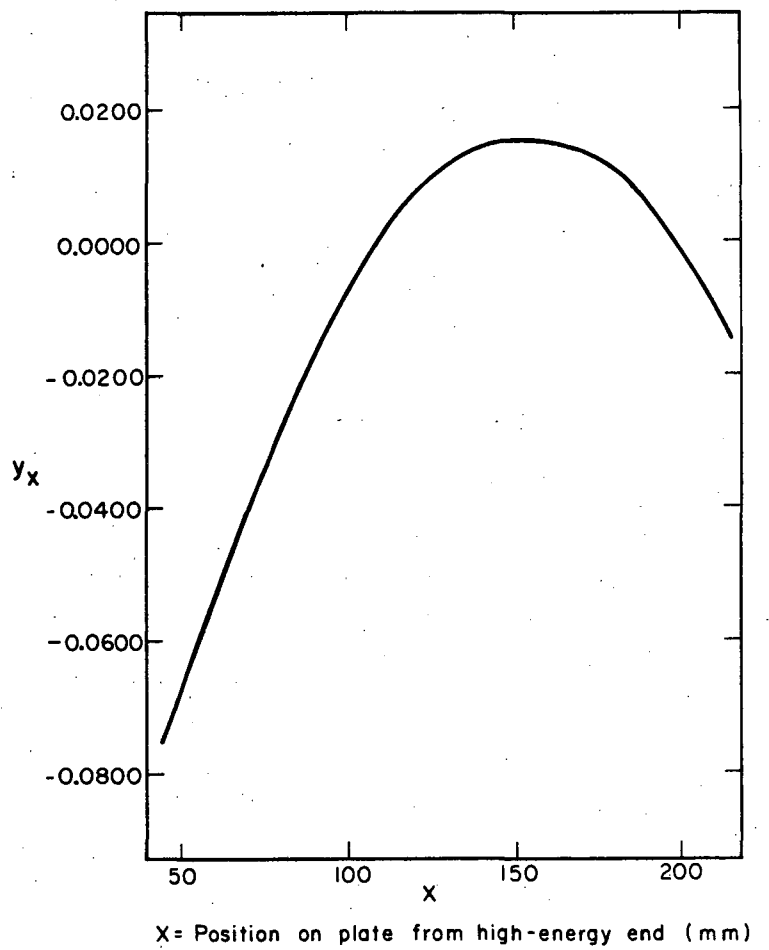
from the source.<sup>9</sup> In this way lower intensity alpha groups could be seen than would otherwise be possible.

The resolution of the spectrograph depends almost entirely on the slit and baffle conditions used and also on the condition of the sample (this latter point will be considered in Section D-2). At baffle openings greater than  $\pm 5$  cm the peaks throughout the whole region of the plate are severely distorted by extensive high-energy tailing. At  $\pm 5$  cm opening distortion occurs mainly at the ends of the plate--the useful region being restricted to about a 100 mm region in the center. As the baffle opening is decreased the useful region increases so that at  $\pm 1$  cm opening the distortion occurs only at the extreme ends of the plate. Because of the double focusing properties of the spectrograph a virtual image of the source is obtained on the plate. Therefore the narrower the source can be made, the better the resolution. For high resolution (less than 0.1 percent) a slit of .010 inch is used with a baffle opening of  $\pm 2$  cm or less. The limitation of the resolution is imposed by the amount of activity in the source and its specific activity. Since only a limited amount of activity could be produced in this research (in the case of  $U^{229}$ ) or the specific activity was low ( $U^{233}$ ) fairly wide slits and baffle openings had to be employed so the resolution ranged from 0.1 to 0.3 percent.

In order to find the separation between peaks, the positions of the peaks were taken as the midpoints at half their maximum height, which has been shown to be <sup>the</sup> most reliable method. Calibration curves have been obtained from alpha emitters of known energy in order to determine the energy separation between groups, and the following equation developed:

$$\frac{E \text{ (kev)}}{H^2} = 14.0486 - 0.0091330x + y_x, \quad (2)$$

where H is the effective magnetic field in megacycles, x is the position on the plate in mm from the high energy end, and  $y_x$  a correction term is shown in Figure 2 plotted as a function of x. The quantity  $H^2$  is usually determined from the energy and position of a standard peak in the spectrum. Knowing this, the energies of all the other groups in the spectrum can be calculated.



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Fig. 2. Calibration curve for alpha particle spectrograph.

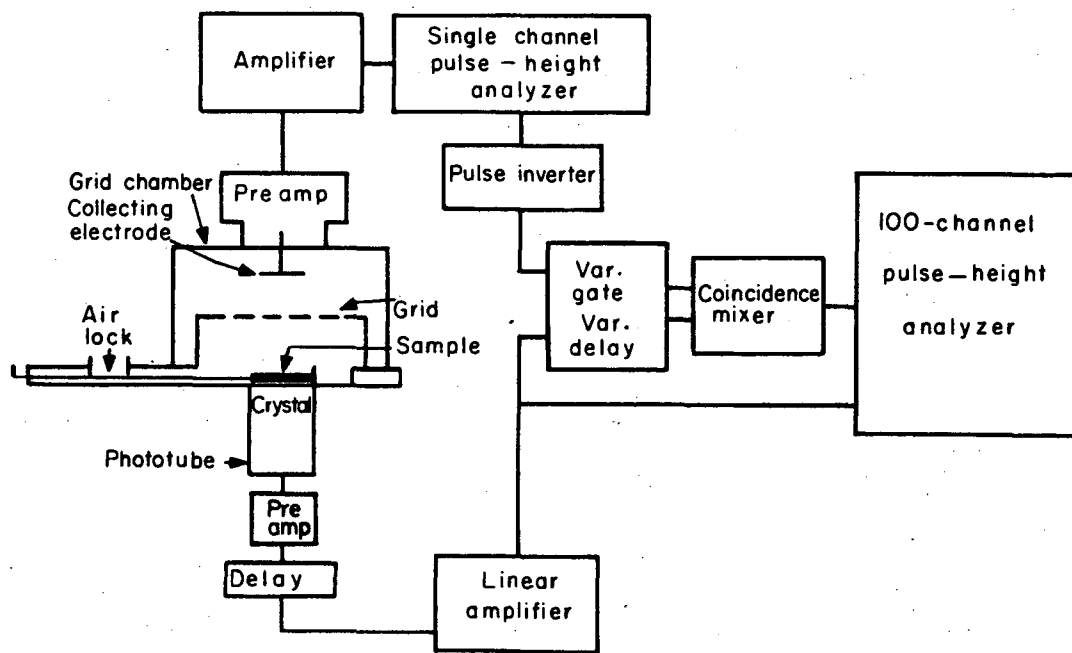
It should be mentioned that the receiver containing the plate can be replaced by a zinc sulfide screen masked except for a vertical slit and coupled through a short light pipe to a photomultiplier tube. The source holder is constructed with a beryllium window thus making possible the detection of gamma rays by a sodium iodide scintillation assembly. Coincidences can then be run by focusing a certain alpha group on the zinc sulfide screen and observing the gamma rays on a multi-channel pulse-height analyzer. Angular distributions can also be obtained by varying the angle of the gamma detector. Neither of these features were used because the source strength was too weak.

For short-lived activities it was necessary to get the source into the vacuum system rapidly. The source holder assembly just described was replaced by one having a vacuum tight valve to isolate a small chamber which could be opened to air while maintaining the rest of the spectrograph under vacuum. The sample was mounted on a brass plate and a vacuum tight seal provided by means of an "O" ring. When the brass plate was in position, the small chamber could then be evacuated quickly and opened to the chamber proper. The time involved for mounting the sample and starting a run was less than one minute; this was used to obtain the alpha spectrum of the Th<sup>225</sup> family.

## 2. Alpha-particle ionization chamber

This ionization chamber utilized a grid to shield the collecting electrode from positive ions.<sup>10</sup> The sample holder served as one electrode and was maintained at a potential of 2.5 kilovolts negative and was insulated from the rest of the chamber. The grid, consisting of 0.004 inch diameter parallel wires 0.035 inches apart, at a distance of 5.5 cm from the sample electrode was at a negative potential of 1.5 kilovolts. The collecting electrode was at a distance of 2.5 cm from the grid and was grounded (a schematic is shown in Figure 3 in conjunction with a coincidence circuit).

The sample could be introduced into the chamber by sliding the sample electrode out of the ionization chamber into a small air lock which was then sealed off from the main chamber by an "O" ring. (Contact with the 2.5 kilovolt supply was automatically broken as the electrode was slid out.) This small compartment could then be let down to atmospheric



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Fig. 3. Schematic drawing of alpha particle grid chamber and block diagram of accompanying coincidence circuit.

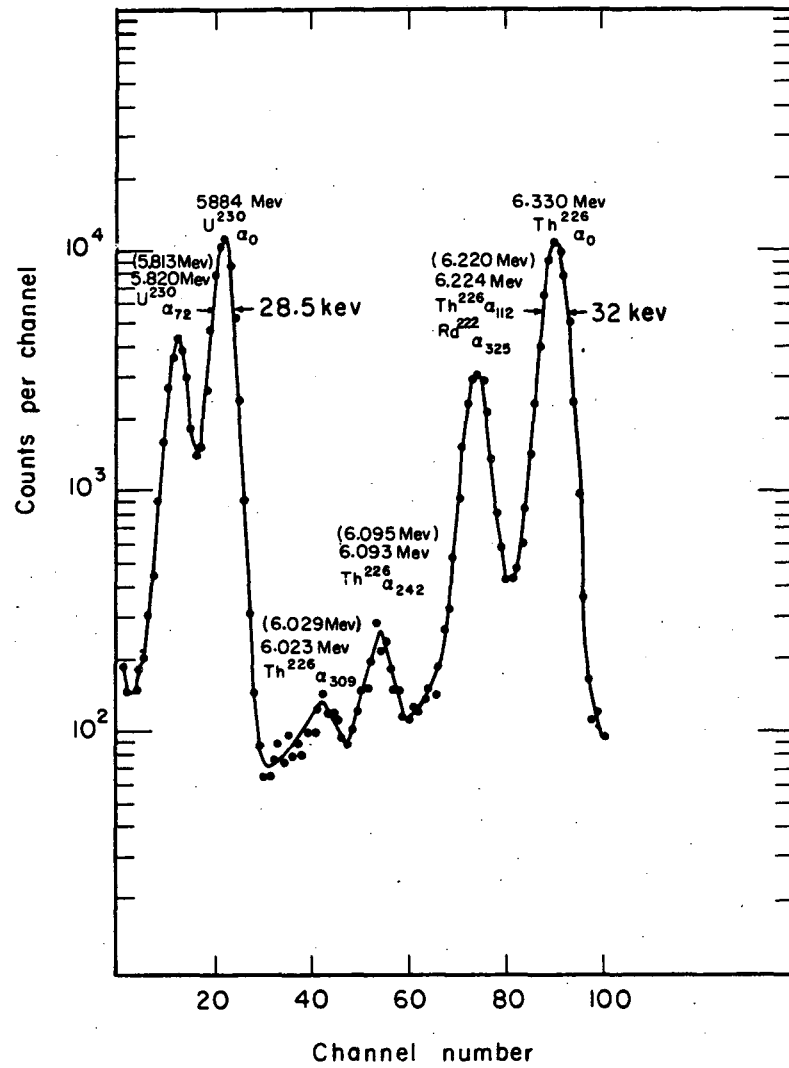
pressure to introduce the sample, and then be evacuated, filled with the gas mixture, and slid into position again without exposing the whole chamber to air. A gas mixture of 93 percent argon and 7 percent methane was used; this provided a short collection time of the electrons produced by ionization and saturation could be achieved at reasonable field strengths.<sup>11</sup> The chamber was usually operated at a pressure of 1.8 atmospheres with a flow rate of 50 cm<sup>3</sup> per minute.

Since this chamber was to be used for coincidence work (circuit described in following section), a 0.010 inch beryllium window was provided in the sample electrode, which was located 1 cm above the floor of the chamber, and a 0.030 inch beryllium window in the chamber floor. A scintillation detector could then be placed underneath the chamber within 1 cm of the sample.

The collecting electrode was coupled to the grid of the first tube of the preamplifier which set directly on top of the grid chamber. Microphonics arising at the input were reduced by mounting the whole assembly on top of a 2 inch sponge rubber strip. The output pulse from the preamplifier had rise and fall times of 2μ second and was about 4μ second long. This 1 volt pulse with a signal to noise ratio of 200 was fed into the main amplifier. In the first part of the amplifier the pulse was amplified and sent into the discriminator part which clipped off the lower portion of the pulse and amplified the rest to the requisite amount. The pulse from here was then sent into a 100-channel analyzer.

All the sources used in the grid chamber were prepared by vacuum sublimation (described in Section 3-D) and were collimated with either a metal ring 3 mm high and 23 mm in diameter<sup>12</sup> or electroformed nickel screen<sup>13</sup> 0.004 inch thick with 80 or 100 holes per inch. The mesh was used to greatly reduce peak distortion in those cases where a highly converted transition was coincident with an alpha particle. The transmission was about 30 percent for the ring collimator and about 3 percent for the mesh collimator.

An example of the resolution which was obtained is shown in the spectrum of U<sup>230</sup> and Th<sup>226</sup> (Figure 4) where the half-width of the U<sup>230</sup> α<sub>0</sub> peak is 28.5 kev, and that of the Th<sup>226</sup> α<sub>0</sub> is 32 kev. This sample was collimated with the 100 count mesh and one can see the U<sup>230</sup> α<sub>72</sub> peak



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Fig. 4. Alpha particle spectrum of  $U^{230}$  and  $Th^{226}$ . (Energies in parentheses are from literature).

clearly resolved, but shifted toward the  $\alpha_0$  peak by 4 kev indicating that the conversion electron effect is greatly reduced (without the mesh collimator it appears only as a shoulder on the  $\alpha_0$  peak) but not eliminated. By more stringent collimation this could probably be improved.

In order to estimate the contribution to the peak width caused by chamber, source, ionization straggling and amplifier noise, the standard deviation of the alpha peak (the half-width is equal to 2.358) was compared with that produced by introducing pulses into the pre-amplifier by a mercury relay pulse generator. The  $U^{230}$   $\alpha_0$  peak having a standard deviation of 12.1 kev was taken since the alpha groups of  $Th^{226}$  were broadened slightly because some of the  $Th^{226}$  are recoiled into the plate from the  $U^{230}$  alpha decay. The "jitter" in the pulses from the pulse generator was observed to be very small and so by feeding these into the preamplifier a standard deviation for amplifier noise of 9.4 kev was obtained. The contribution to the standard deviation from the three types of straggling is 7.7 kev. Since ionization straggling alone has been estimated to be 7 - 9 kev,<sup>14</sup> then it is seen that the contribution from source and chamber straggling is negligible. Therefore, if the resolution of the system is to be substantially improved, the preamplifier noise (which depends almost entirely on the noise level of the first amplifier tube) must be reduced. This could only be done by obtaining a lower noise tube or perhaps redesigning the preamplifier circuit. However, it is seen that this resolution is adequate to resolve the fine structure occurring in even-even alpha emitters by referring to the four alpha groups of  $Th^{226}$ , which are clearly exhibited in the spectrum (Figure 4).

The relatively high transmission of this instrument compared to that of the spectrograph with a loss in resolution of only a factor of five made the experiments with the  $U^{228}$  family possible as well as many coincidence experiments with the  $U^{229}$  family and  $U^{233}$ .

#### B. Gamma-ray spectroscopy

Gamma-ray spectra were obtained with scintillation spectrometers which were 1 1/2 by 1 inch or 3 by 3 inch sodium iodide (Tl activated) crystals mounted in the conventional way. Nearly all the gamma-ray

spectra were obtained by employing coincidence techniques; i.e., gamma-rays in coincidence with all alpha particles, with alpha particles of a particular energy, or gamma-rays of a particular energy. A block diagram of the basic coincidence circuit used is shown in Figure 3 where the detectors are the grid chamber and a scintillation crystal. In this particular arrangement the signal pulses from the gamma detector are passed through a delay and then amplified. The output pulse from the amplifier is branched, one branch going to channel one of a variable gate-variable delay unit while the other branch is fed into the signal input in the 100-channel pulse-height analyzer. The pulse is recorded only if the analyzer is externally triggered simultaneously by a gate pulse. The gate pulse is obtained in this case by passing the alpha pulse from the amplifier through a single-channel analyzer and pulse-inverter into channel 2 of a variable gate-variable delay unit. After shaping and delay adjusting, the pulses from both channel 1 and channel 2 are mixed and used as the trigger pulse for the 100 channel analyzer. The circuit was usually operated at a resolving time of  $2.5\mu$  second. With this type of circuit, gate pulses could be selected from either channel separately or from coincidence pulses between the two channels.<sup>15</sup> Thus singles spectra could be run by externally triggering the pulses. This avoids any uncertainty which might be introduced in energy calibration and comparison of singles spectra with coincidence spectra by switching from externally to internally triggered spectra. For gamma-alpha coincidence studies the roles of the detectors were reversed with obvious changes made in the circuitry.

For gamma-gamma coincidence studies the grid chamber and amplifier were replaced by another sodium iodide scintillation assembly with accompanying linear amplifier. For total alpha-gamma coincidence studies, the alpha detector was a photomultiplier tube sprayed with zinc sulfide. Since a higher geometry could be achieved for the gamma detector with this arrangement, it was preferred over the grid chamber. (A single-channel analyzer was not used in this case.)

For all spectra obtained, chance coincidence corrections have been made and intensities of all gamma rays have been corrected for escape peak losses and counting efficiencies.

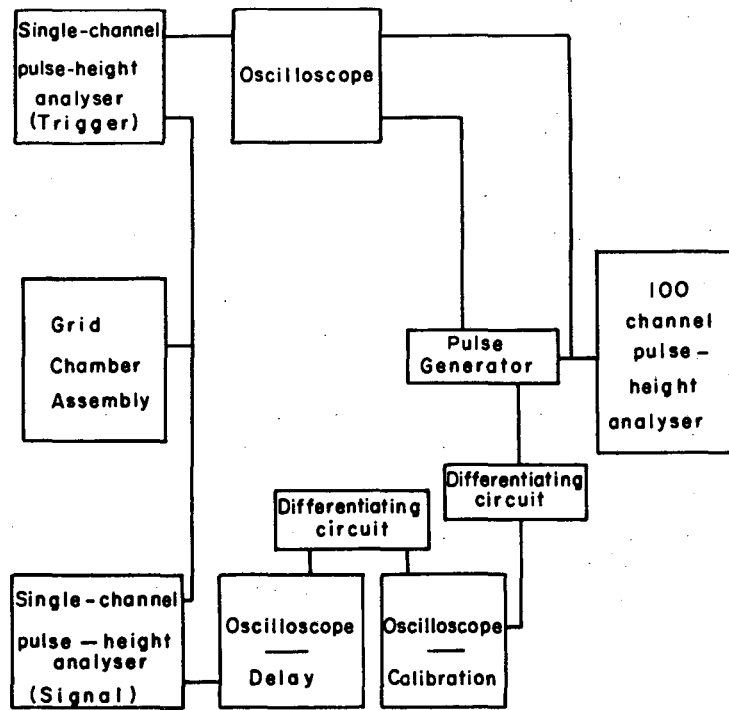
### C. Half-Life Determinations

Since the half-lives of the activities measured were appreciably less than one second (with the exception of  $U^{228}$ ), the following electronic system was developed to obtain the information for  $Rn^{216,217,218}$ , while a fast moving tape method was used for  $Ra^{220}$  (Section C-2).

#### 1. Electronic System

A block diagram of the electronic system is shown in Figure 5. The alpha pulses originating from the grid chamber were amplified in the preamplifier and main amplifier (grid chamber assembly). At the amplifier output the pulses were branched and fed into two single-channel analyzers. One was set to pass radium alpha pulses (trigger) and the other set to pass radon alpha pulses (signal). The trigger pulse was used to trigger the sweep of a Tektronix-type 541 oscilloscope from which a sawtooth pulse was taken and passed into a mercury relay pulse generator. This made the amplitude of the output pulse occurring at time  $t$  after the time of the trigger pulse,  $t_0$ , proportional in amplitude to the height of the sawtooth pulse at time  $t$ . In addition, this pulse generator was modified to give a pulse only when externally triggered. The signal pulses (from the other single channel analyzer) were passed through one oscilloscope which was used as a 2 millisecond delay (to compensate for delay of the mercury relay), and through a second oscilloscope which was used for calibration. After going through a differentiating circuit, these were used to trigger the pulse generator. This time-to-pulse-height converter system supplied pulses to a 100 channel pulse-height analyzer, the height of the pulses being proportional to the time delay between the trigger pulse and the signal pulse. This arrangement could be used to measure half-lives from a few minutes to  $10^{-4}$  seconds. The spectrum obtained was that of a typical decay curve complete with a tail due to chance coincidence counts which was greatly reduced by selecting only the alpha particles of interest (by means of the single-channel analyzers).

The system was calibrated in the following way: both single channel analyzers were set to pass the same alpha group giving pulses simultaneously through both parts of the circuit. Then by varying the sweep time (and thus the delay) of the calibration scope on the trigger side, the trigger pulse was varied in time in such a way that it would fall successively on the calibrated markings as observed on the scope on the gate side. Simultaneously the pulses were fed to the 100-channel



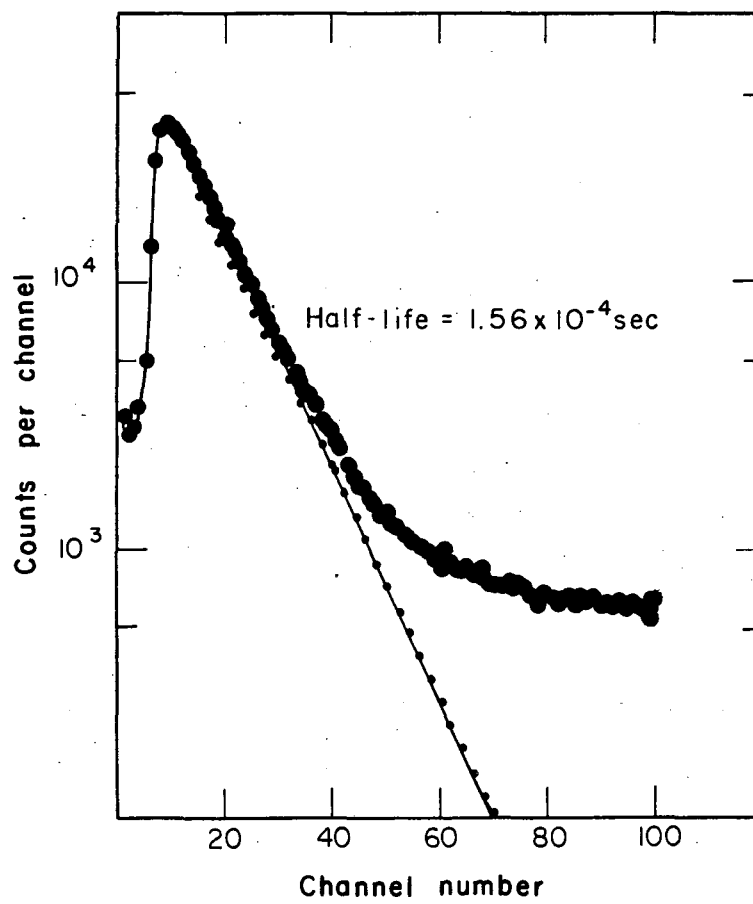
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Fig. 5. Block diagram of time-to-pulse-height converter system.

pulse-height analyzer. A calibration curve was thus obtained relating the channel number to the time from the start of the oscilloscope sweep as observed on the gate oscilloscope. Such a curve, using the mercury relay pulse generator, was linear for all sweep speeds within the range of operation.

To check the operation of the system, the well known half-life of  $\text{Po}^{214}$  was measured and the curve of this is shown in Figure 6. The literature values are  $1.64 \times 10^{-4}$  and  $1.58 \times 10^{-4}$  second; the values obtained from this curve and from another run were  $1.56 \times 10^{-4}$  and  $1.66 \times 10^{-4}$  second.

It was found that the delay of the mercury-relay pulse generator was sensitive to changes in the count rate of the signal pulse above a few hundred counts per minute. As the count rate was increased the delay would increase and eventually reach a maximum of 4 milliseconds at a count rate of about 1500 c/m. This was not a serious limitation as long as the half-lives of the parent activities were comparatively long so that one could keep the count rate low and constant. However, in the case of  $\text{Rn}^{216}$  these conditions could not be met. The  $\text{Rn}^{216}$  had to be studied in equilibrium with  $\text{U}^{228}$  which is a 9-minute activity. This meant that in order to obtain reasonably good statistics, the count rate would have to be fairly high and also would change by at least one order of magnitude throughout the run. This would certainly affect the delay of the pulse generator which would be fatal to the determination of the half-life estimated to be about  $10^{-4}$  second. For this measurement, the mercury-relay pulse generator was replaced by a unit consisting essentially of a multivibrator whose pulse output was of constant amplitude and a discriminator. The same arrangement (Figure 5) was used with the exception of the delay scope on the trigger side which was no longer needed since this unit had no appreciable delay. The technique of operation was exactly the same as that previously described. The sawtooth pulse would increase the voltage on the discriminator which would subtract from the pulse output of the multivibrator; thus the later in time a trigger pulse would arrive after the gate pulse (the start of the sweep) the smaller would be the output pulse. This system could be used to measure half-lives ranging from several minutes to  $10^{-5}$  second and could take count rates up to 500 c/m very easily. However it suffered from the fact that it was not linear. This was not too serious and could be corrected. The system was



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Fig. 6. Delay-coincidence curve of  $\text{Po}^{214}$ .

checked by measuring  $\text{Rn}^{217}$  which was measured with the previous system and the two values found to agree within 5 percent. This was then used to measure the half-life of  $\text{Rn}^{216}$ .

## 2. Tape Method

The electronic systems described above could not be used to measure the half-life of  $\text{Ra}^{220}$  (estimated half-life of 0.03 second). In order to get fairly good statistics, a comparatively high count rate would have to be used because the experiment was limited by the 9-minute half-life of the grandparent. Under these conditions the chance coincidence rate would be prohibitively high. A fast moving tape method was therefore devised to make the measurement; a schematic drawing of this is shown in Figure 7. A 1-inch wide by 0.002-inch thick stainless steel tape made into a belt about 30 inches long was driven by an electric motor at the rate of 85-inch per second. The belt was held in position by the tension exerted by its pulling against two stainless steel spools, one attached to the shaft of the motor and the other to a freely rotating shaft. About 1/2 inch below the tape was a row of six alpha detectors, consisting of zinc sulfide sprayed photomultiplier tubes with their accompanying assemblies. The pulses from each of these tubes were fed through linear amplifiers into a bank of six scalers. The zinc sulfide screens were masked off so that the active area of each screen was 1 inch square. The spacing between the active area of each detector was 2.5 inches. The activity was recoil-collected on the tape by placing a  $\text{U}^{228}$  sample (prepared by vacuum sublimation, Section D-2) about 1/4 inch beneath the tape and applying a potential of 1200 volts between this and an electrode 1/4 inch above the tape. The sample was about 1 inch from the first detector.

After making a counting efficiency correction for the counters, a decay curve was obtained. From a knowledge of the speed of the tape and the distance between the detectors, the detector position was related to time delay.

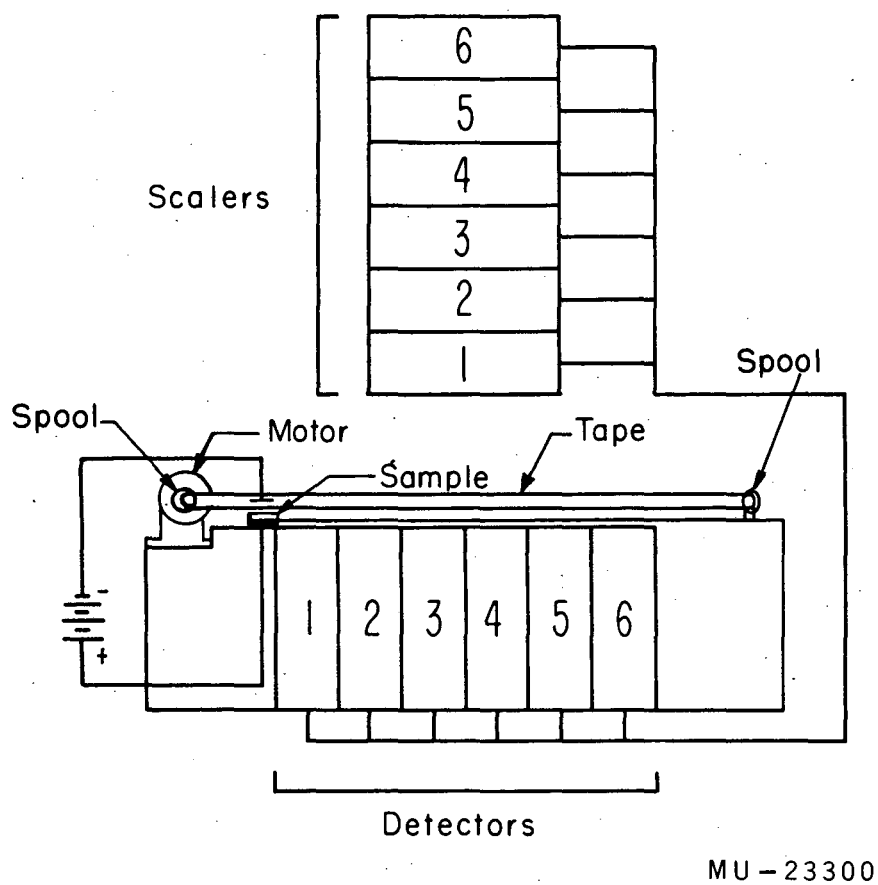


Fig. 7. Schematic drawing of arrangement for half-life measurement with moving tape.

## D. Production and Purification of Active Materials

### 1. Bombardment Procedures

The light-uranium isotopes studied were produced by bombardment of  $\text{Th}^{232}$  with the internal helium ion beam of the 184-inch synchrocyclotron. The target was placed at the innermost accessible radius (45 inches) in order to obtain the minimum energy of 300 Mev. The target, consisting of two or three strips of 0.010 inch thorium metal measuring about 1 by 0.4 inches, was bombarded on end. The target was bombarded at full beam intensity (roughly 0.1 microampere) for a time equal to 1 - 2 half-lives of the activity desired.

An alternate method to produce  $\text{U}^{228}$  free from the other uranium isotopes was attempted. This involved bombarding uranium foil, using the same technique as described above, to make  $\text{Pu}^{232}$ . The  $\text{U}^{228}$  could have then been recoil-collected yielding a sample completely free from contaminants. This also had the advantage of having a longer time to work with the uranium for each bombardment, since the half-life of  $\text{Pu}^{232}$  is 27 minutes. However, the extremely low yields of  $\text{Pu}^{232}$ , resulting from bombardment of both  $\text{U}^{235}$  and  $\text{U}^{238}$ , made this approach impossible.

### 2. Chemical Separation

By far, the largest part of the total cross section in a 300 Mev helium ion bombardment of thorium, goes into fission, thus greatly decreasing the yield of  $\text{U}^{228}$  and  $\text{U}^{229}$ . Therefore the uranium must be separated from essentially every other element in the periodic table as well as from relatively large amounts of target material. Furthermore, because of the high radiation level, it is necessary to perform most of the operations behind adequate shielding where mechanical tongs are used to reduce the radiation dose on the hands. Under these conditions, complicated or delicate operations are tedious and extremely time consuming and must be avoided, considering the half-lives of the products involved. Since the bombardment yield is low, the chemical yield must be high to make the experiments possible. The requirement of a mass free sample for alpha spectroscopic measurements further complicates the problem. From a consideration of these requirements, a chemical procedure was developed using methods of ion exchange and solvent extraction.

The thorium metal was dissolved in a solution of concentrated  $\text{HCl}$  - 0.2 M  $\text{HF}^8$  to which a few drops of concentrated  $\text{HNO}_3$  had been added

to oxidize the uranium to the (VI) state. Dissolution was complete within one minute with only a small amount of a white solid material (presumably  $\text{ThO}_2$ ) remaining. This solution was loaded into a column (30 mm in height and 5 mm in diameter) packed with Dowex A-1 anion exchange resin. It was forced through the column under pressure at a flow rate of 2 drops per second. Under these conditions the uranium as a uranyl chloride complex, is adsorbed on the column while the other alpha activities (with the exception of polonium and protactinium) as well as many of the fission products pass through with little or no adsorption.<sup>16</sup> Several column volumes of 6 M HCl were then passed through the column improving the decontamination and also ensuring the removal of the target material in preparation for the next step. About two column volumes of 7 M HCl - 0.5 M HF were passed through to ensure the complete removal of protactinium<sup>17</sup> while the uranium still remained strongly adsorbed.<sup>18</sup> The resin was then washed with 1 column volume of 4 M HCl to remove the fluoride ion in preparation for an ether extraction,<sup>19</sup> and finally the uranium was eluted by passing through 1 ml of 0.7 M HCl. At this point the beta and gamma activity had been reduced by two orders of magnitude but further chemistry was needed to reduce it still more. At this point the procedures for  $\text{U}^{228}$  and  $\text{U}^{229}$  became slightly different.

In the case of  $\text{U}^{229}$  the 1 ml solution was evaporated to dryness and the activity then taken up in 1 M  $\text{HNO}_3$  and transferred to a test tube containing a 10 M  $\text{Ca}(\text{NO}_3)_2$  solution making the solution about 0.05 M in  $\text{HNO}_3$ . Two volumes of ether were added and the system stirred for about 2 minutes. The ether phase (containing the uranium) was pipetted off and washed with a saturated  $\text{NH}_4\text{NO}_3$  - 0.1 M  $\text{HNO}_3$  solution and then transferred to a beaker containing 2 drops of 4 M HCl and evaporated to dryness. A few drops of concentrated HCl were then added and the solution passed through another anion exchange column (3 mm in diameter, 10 mm high) washed with 6 M HCl and eluted with 0.7 M HCl. The eluant was collected on a Pt plate and evaporated to dryness. This chemistry reduced the beta and gamma activity another factor of a hundred and produced a sample which was fairly clean but not completely mass free. The sample was then vacuum sublimed. The chemical yield for  $\text{U}^{229}$  was about 70 percent and the time involved from the end of bombardment was about 75 minutes. The average bombardment yield was  $10^5$  alpha counts per minute.

For  $U^{228}$ , the 1 ml eluant was collected in a test tube containing a saturated  $Al(NO_3)_3$  - 0.1 M  $HNO_3$  solution with excess  $Al(NO_3)_3$  added to saturate the 1 ml eluant, and an equal volume of ether. This was stirred and then the ether phase was pipetted off and washed with a saturated  $NH_4NO_3$  - 0.1 M  $HNO_3$  solution. The ether phase was evaporated (time did not permit another column separation) in a small beaker leaving only the  $HNO_3$  extracted by it containing the activity. This was then vacuum sublimed. The chemical yield for the whole process was about 60 percent and the time involved from cyclotron shut-down was about 17 minutes. An average of  $5 \times 10^4$  alpha counts per minute was obtained.

All of the requirements that the chemical procedure must satisfy has been met except the requirement of a thin, uniform, mass free sample. This was mandatory for obtaining the resolution possible with the alpha spectrograph, and was also of importance for the grid chamber; it was also essential for high recoil collection efficiency of the daughter activities which provide a very clean separation from the parent activity. The process of vacuum sublimation was employed to achieve this. The uranium activity was evaporated from the hydrochloric or nitric acid solution onto a tungsten or tantalum filament about 1/2 inch long. The filament was folded to make a "V" in order to "focus" the activity into a line for the spectrograph. The system, enclosed by a bell jar, was then evacuated to a pressure of 0.5 micron. The filament was preflashed to as high a temperature as possible without losing any uranium, thus removing as many of the more volatile impurities and extraneous mass as possible. A 0.002 inch plate, either platinum or aluminum was placed a fixed distance above the filament, and after re-evacuation, the filament flashed to white heat to sublime the activity to the plate. This sublimation occurs within a small temperature range and so by controlling the conditions, a partial separation could be achieved.

This feature of fractional sublimation was greatly needed especially in the case of  $U^{228}$ , because even after chemistry, an appreciable amount of beta-gamma activity remained (probably because of the rapidity with which the chemistry had to be done) which would have rendered coincidence work impossible.

After the  $U^{229}$  had decayed away, the  $Pa^{229}$  which was formed was taken off the plate and the solution made 10 M  $HCl$ . It was contacted

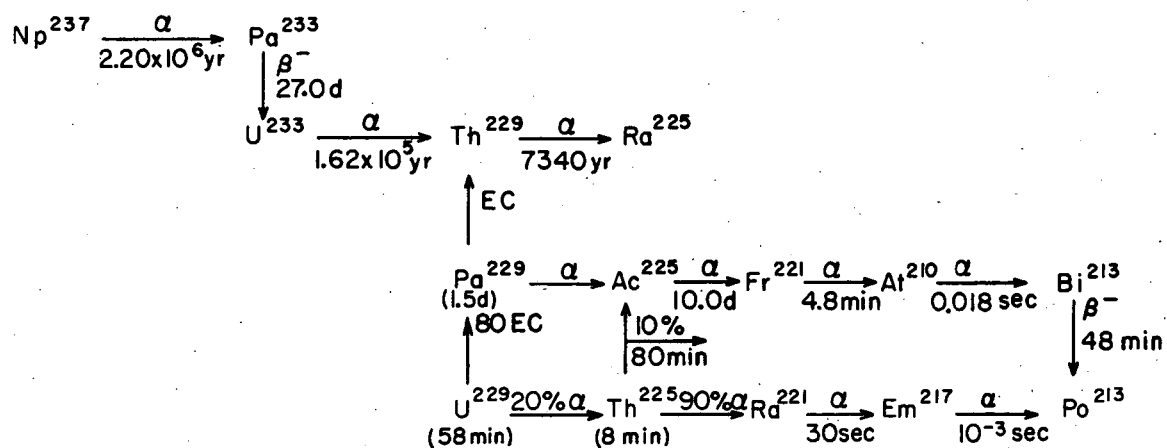
with an equal volume of di-isopropyl ketone (DIPK) into which the  $\text{Pa}^{229}$  was extracted. The DIPK was then contacted with a 2 M HCl solution into which it was back extracted.<sup>10</sup> The solution was then made 10 M in HCl and passed through a short anion exchange column. The  $\text{Pa}^{229}$  was eluted with 7 M HCl - 0.5 M HF solution and evaporated to dryness on a silver plate.<sup>17</sup> This was used for gamma-gamma coincidence studies.

In the case of  $\text{Pu}^{232}$ , only column separations were employed. The  $\text{U}^{235}$ -foil target was dissolved in concentrated HCl and then converted to the nitrate. To this 2 M  $\text{HNO}_3$  solution was added ferrous ion and hydrazine dihydrochloride to make the final concentration 0.005 M in ferrous ion and 0.1 M in hydrazine. The solution was heated for 5 minutes to reduce the plutonium to the (III) state. The plutonium was then oxidized to the (IV) state by making the solution 0.1 M in sodium bromate and heating for 1 minute.<sup>21</sup> Enough nitric acid was added to make the solution 8 M and then it was forced through an anion exchange column (5 mm in diameter, 30 mm in height). Under these conditions the uranium and many fission products passed through with the plutonium remaining strongly adsorbed.<sup>22</sup> The column was washed with 4 M  $\text{HNO}_3$  and then the plutonium eluted with 2 M HCl. The solution was made 10 M in HCl and passed through a second anion exchange column (10 mm by 4 mm) jacketed for operation at 87° C. After washing with more 10 M HCl, the plutonium was eluted by the addition of 10 M HCl - 0.1 M HI reducing it to the (III) state, which is not adsorbed on the resin. This sample was then vacuum sublimed. Sample preparation was complete about 60 minutes after bombardment. The amount of  $\text{Pu}^{232}$  produced was extremely small.

### III. RESULTS

#### A. Uranium-229 Series

The uranium isotope  $\text{U}^{229}$  has a half life of 58 minutes and decays principally by electron capture with about 20 percent alpha branching. The alpha decay produces a series of short-lived daughters, which is collateral to the neptunium ( $4n + 1$ ) family, and joins the main family at  $\text{Po}^{213}$  (Figure 8).<sup>23,24</sup> The alpha particle energies<sup>23</sup> were determined with an ionization chamber to be 6.42, 6.57, 6.71, and 7.74 Mev respectively for  $\text{U}^{229}$ ,  $\text{Th}^{225}$ ,  $\text{Ra}^{221}$ , and  $\text{Rn}^{217}$ . This summarizes all the data which have been previously reported.



MU-23301

Fig. 8. Neptunium family and collateral  $U^{229}$  family.

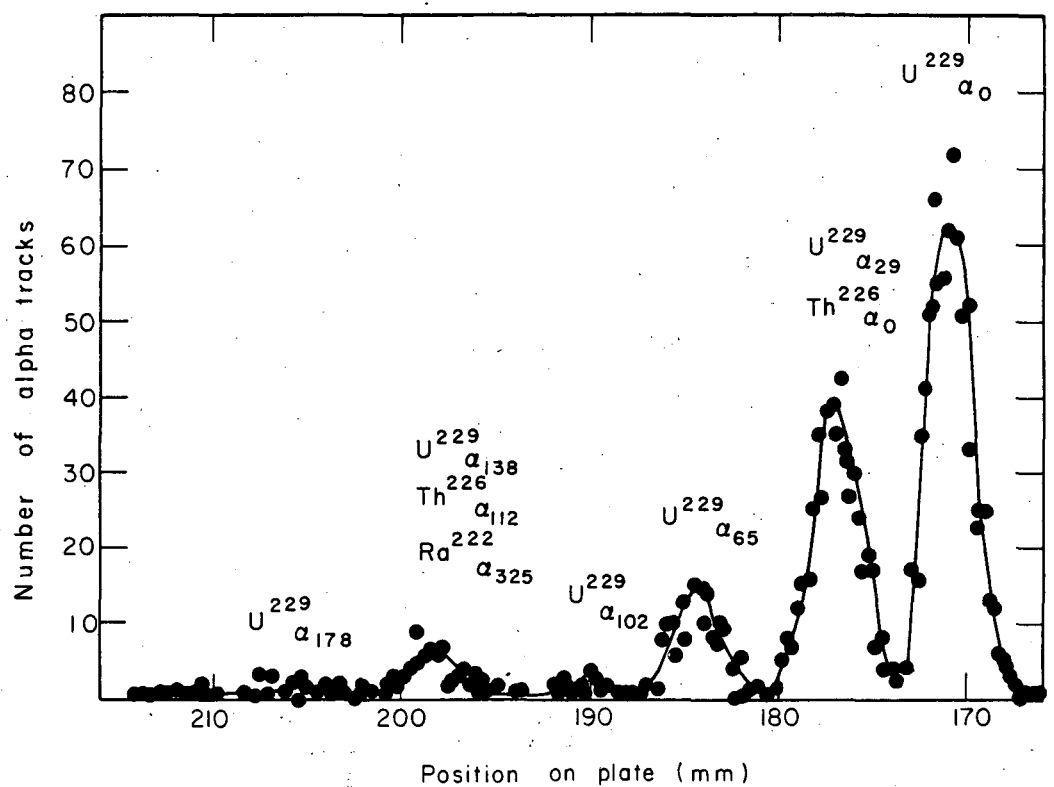
# 1. Uranium-229

## a. Alpha Spectrum

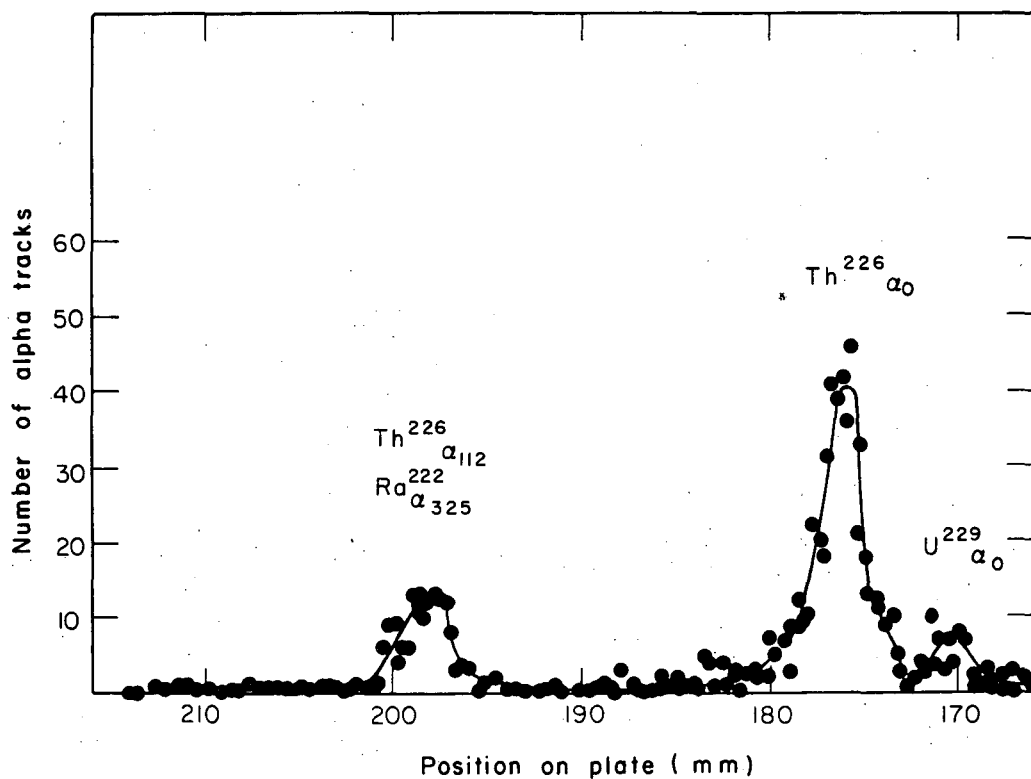
Because of the short half-lives of the daughters, these rapidly grow into equilibrium with the parent activity after separation, so that the alpha spectrum of  $U^{229}$  includes the daughters,  $Th^{225}$  and  $Ra^{221}$ . The  $Rn^{217}$  and  $Po^{213}$  which are also present do not interfere because their alpha particle energies are outside of the range of interest.

Figure 9a covers that portion of the spectrum containing the alpha groups which will be shown to be due to  $U^{229}$ . Figure 9b is the spectrum of the same region taken after the  $U^{229}$  had essentially decayed out (the exposure time was about twice as long). By comparing the two spectra, the peaks due to the  $U^{230}$  family could readily be assigned. The two spectra were normalized with respect to length of exposure and it was determined that  $U^{229}$  had peaks which coincided in position with  $Th^{226}\alpha_0$  and  $Th^{226}\alpha_{112}$  (and  $Ra^{222}\alpha_{325}$ ). Besides the comparatively intense groups, there is an indication of groups at the 190 and 206 mm positions (Figure 9a) although the statistics are not very convincing. The existence of these groups was proved in an exceptional run having about 5 times the activity. The region of interest is shown in Figure 10; a quantitative comparison of this spectrum with one obtained after the  $U^{229}$  had decayed strongly suggested the existence of the two groups, (indicated as  $U^{229}\alpha_{102}$  and  $U^{229}\alpha_{178}$ ).

With six groups in this energy interval assigned to the  $U^{229}$  series, it was now necessary to determine which of these belonged to  $U^{229}$  itself. The spectrum due to  $Th^{225}$  and its daughters was measured on samples by collecting recoils from a thin  $U^{229}$  source. Recoils were collected in air for 8-minute intervals on a 10-mil platinum wire maintained at 1200 volts and inserted rapidly into the spectrograph through the special air lock already mentioned. In order to build up a sufficient number of alpha tracks on the photographic plate the source was replaced many times after fresh collection of recoils. The spectrum obtained is shown in Figure 11a. Another plate was inserted after members of the  $U^{229}$  series had decayed in order to obtain separately the longer-lived products collected as recoils. This spectrum is shown in Figure 11b. The arrows in Figure 11a mark the positions where groups from  $U^{229}$  should appear. It is obvious that the groups designated as  $\alpha_0$  and  $\alpha_{65}$  which

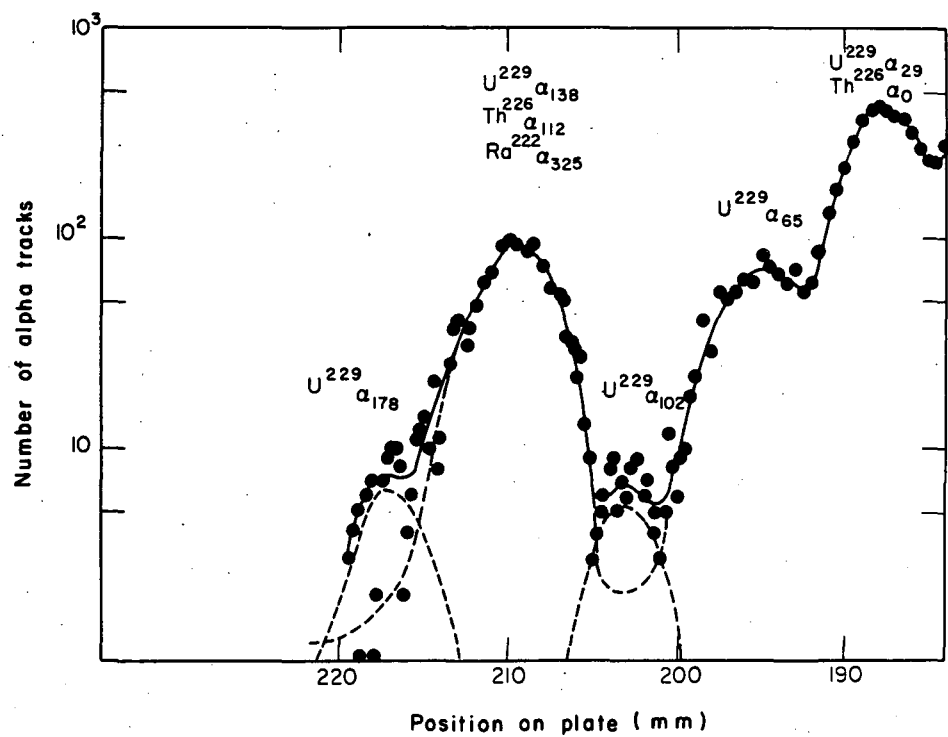


MU-23302



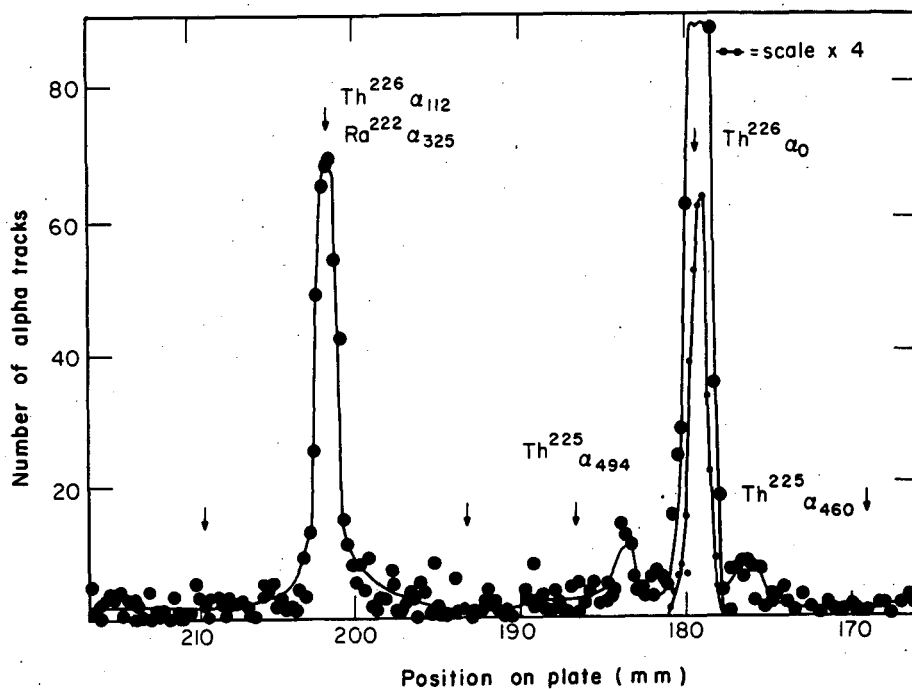
MU-23303

Fig. 9. Alpha spectrum of  $U^{229}$  taken with alpha particle spectrograph.  
 a. Starting 75 minutes after end of bombardment.  
 b. Starting 5 hours after end of bombardment.

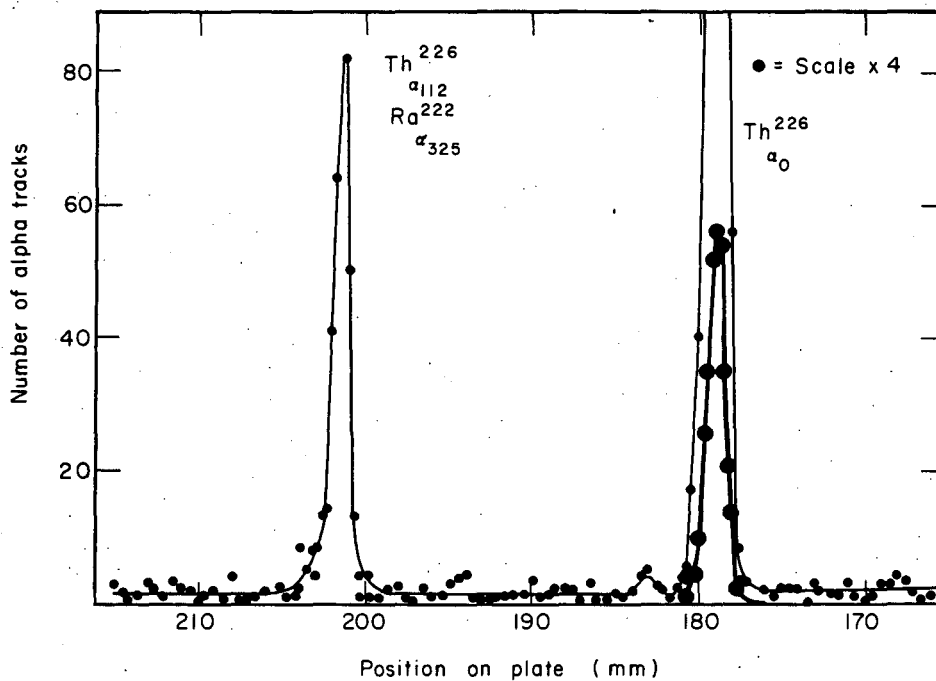


MU-23304

Fig. 10. Alpha spectrum of  $U^{229}$  taken with alpha particle spectrograph showing low intensity groups.



MU-23305



MU-23306

Fig. 11. a. Alpha spectrum of low energy region of  $\text{Th}^{226}$  family taken with alpha particle spectrograph. Arrows indicate positions where  $\text{U}^{229}$  groups occurred (Figures 9 and 10).  
 b. Alpha spectrum of same region after decay of  $\text{Th}^{226}$ .

would appear at the 173 and 187 mm positions in Figure 11a are due to  $U^{229}$  itself.

The peaks occurring at the same position as  $Th^{226}\alpha_0$  and  $Th^{226}\alpha_{112}$  (and  $Ra^{222}\alpha_{325}$ ) were identified as belonging to  $U^{229}$  by a comparison of their intensities to that of the  $Ra^{222}\alpha_0$  group (not shown but present in these spectra at higher energy). The arrows at the 193 and 209 mm positions show the absence of any groups which would be about six tracks tall if they were not due to  $U^{229}$ .

This data is summarized in Table I together with the hindrance factors.\* A search was made for groups of lower energy occurring in the region from 6.18 to 5.884 Mev ( $U^{230}\alpha_0$ ), but none was found within the limit of detection of 1 percent. It will be shown later (Section 2a) that the peaks seen at 176 and 184 mm are due to  $Th^{225}$ .

TABLE I

Alpha Groups of  $U^{229}$

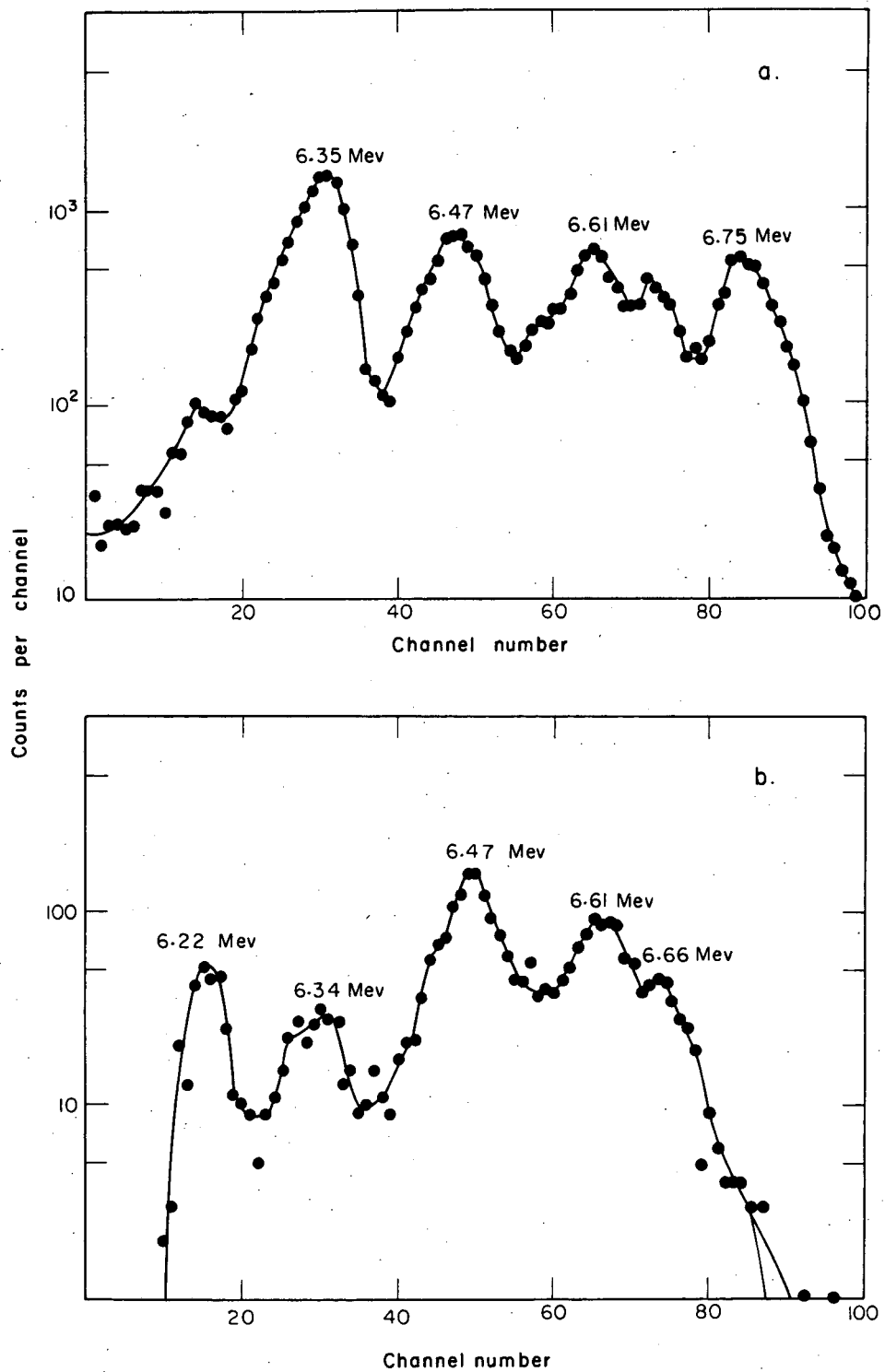
| Alpha-particle energy (Mev) <sup>a</sup> | Excited-state energy (kev) | Abundance % | Hindrance factor |
|------------------------------------------|----------------------------|-------------|------------------|
| 6.355 ± 0.003                            | 0                          | 64          | 1.5              |
| 6.327                                    | 29 ± 2                     | 20 ± 2      | 3.7              |
| 6.292                                    | 65 ± 2                     | 11 ± 1      | 4.6              |
| 6.255                                    | 102 ± 3                    | 1 ± 0.5     | 35               |
| 6.218                                    | 139 ± 3                    | 3 ± 1       | 7.9              |
| 6.180                                    | 178 ± 4                    | 1 ± 0.5     | 16               |

<sup>a</sup> Alpha particle energies are expressed relative to  $Th^{226}\alpha_0$  and  $Ra^{222}\alpha_0$  taken as 6.330 and 6.552 Mev.

#### b. Gamma-Alpha Coincidence Spectrum

In order to determine whether or not the 63 percent group really was the ground state group, all gamma ray energies including L x-rays were gated on and the coincidence alpha spectrum was observed. The singles and coincidence alpha spectra are shown in Figure 12. In the singles spectrum the  $U^{229}\alpha_0$  group (which also contains  $\alpha_{29}$ ) is clearly

\* The hindrance factors in this and succeeding sections were calculated by dividing the experimental half-life by the half-life calculated by the method of Preston.<sup>25</sup>



MUB-640

Fig. 12. a. Alpha spectrum of  $U^{229}$  family taken with alpha particle ionization chamber.

b. Gamma ray-alpha particle coincidence spectrum of  $U^{229}$  family.

seen as the most intense peak in the spectrum. In the coincidence spectrum the peak is plainly absent indicating that the 63 percent group designated as  $U^{229}\alpha_0$  is the real ground state or within the L-electron binding energy of the ground state.

c. Gamma Spectrum

An alpha-gamma coincidence spectrum of  $U^{229}$  in equilibrium with its daughters was obtained. Upon comparison with the coincidence spectrum obtained from the  $Th^{225}$  series (the  $Th^{225}$  was recoil collected from  $U^{229}$ ) the two spectra were found to be identical, thus indicating the absence of any very intense gamma rays associated with the alpha decay of  $U^{229}$ .

In order to make possible the observation of gamma rays of weaker intensity, the following chemical procedure was devised to separate  $U^{229}$  from its daughters. The activity, contained in 2 drops of 6 M HCl, was passed quickly through a very small cation exchange column. Under these conditions the Th and Ra were adsorbed with the uranium passing through. The 2 drops were collected and evaporated on a silver plate. Counting was started 1 minute after elution and lasted for two minutes. This procedure was repeated until a sufficient number of events had been accumulated. Even with this increased sensitivity, no gamma rays were observed in the coincidence spectrum which could be attributed to the  $U^{229}$ . The limit of detection was about 2 percent.

Since no gamma rays were observed, then it was concluded that the excited states populated by the  $U^{229}$  must de-excite by highly converted transition. The L X-ray region of the spectrum was therefore studied. After correcting for the  $U^{230}$  family contribution a comparison of the spectrum of the equilibrated  $U^{229}$  family with that of the  $Th^{225}$  series revealed that there were L X-rays associated with the  $U^{229}$  decay in an abundance of 13 percent (equivalent to 31 percent L-vacancies).

From a consideration of the alpha population to the first two excited states and the lack of gamma radiation, it is obvious that the de-exciting transitions are not E1. This is verified by the rather large proportion of L X-rays associated with the decay. Now for M1 radiation the ratio of L/M conversion is roughly 1.7 in the energy region of interest and for E2 the ratio is about 2.2; for M2 it ranges from 1.8 for a 29 kev transition to 1.2 for a 65 kev transition. Now if each of the excited states decays directly to ground the percentage of L vacancies created

would be  $\frac{2.2}{3.2} (20 + 11) = 21$  percent. This is for the case of an E2 transition for which the number of L-shell conversions is greatest. It is therefore necessary to conclude that the second excited state de-excites mainly to the first excited state.

#### d. Decay Scheme

The alpha decay scheme of  $U^{229}$  is shown in Figure 13. Radiation is shown cascading through the first excited state from the second. This does not imply that the crossover transition is not present but from arguments presented above only the cascade transition is definite. Finally it should be mentioned that because the 1 percent alpha groups were on the limit of detection, their existence is not absolutely certain.

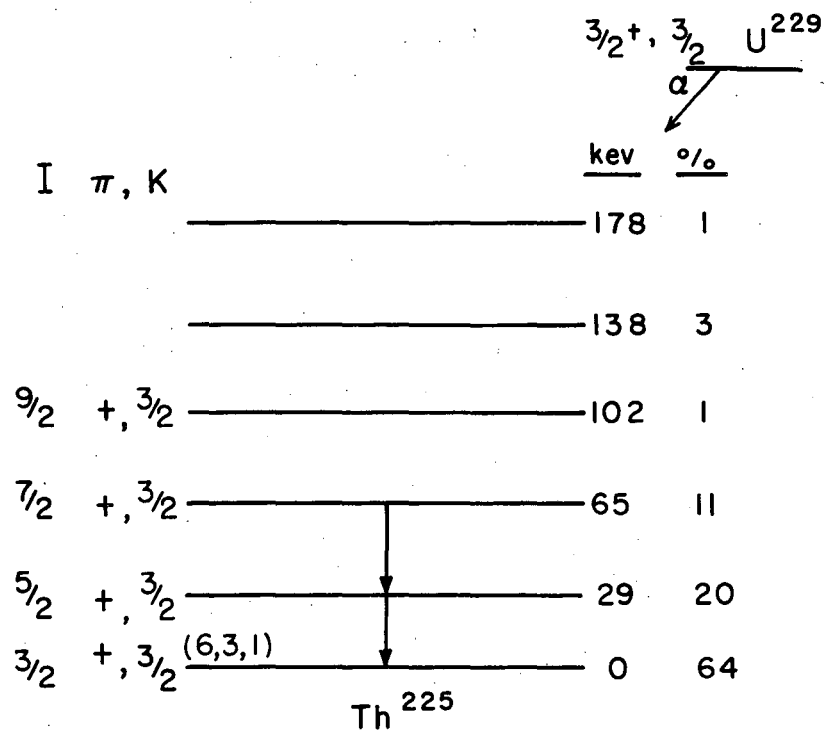
#### e. Interpretation

The low value of the hindrance factor for alpha decay to the ground state of  $Th^{225}$  is characteristic of favored alpha decay whereby the last odd particle remains in the same orbital before and after the decay.<sup>26</sup> The regular variation in intensity and level spacing observed for the first four energy levels is very similar to what is observed in other odd-mass nuclei where these systems of levels are interpreted in terms of rotational excitations based on the same single particle configuration.<sup>27</sup> It is now well established that the Bohr-Mottelson theory is successful in explaining the level schemes in regions of large axial deformation, i.e.,  $A \geq 229$ . What is really the question is how far down below  $A = 229$  the particle states can be reasonably well described by Nilsson's asymptotic quantum numbers<sup>28</sup> and the rotational bands are recognizable. From a study of other nuclei in this mass region, there seems to be no sudden change in the character of the nuclear energy levels. Therefore there is no reason to believe that the character of the levels populated by the  $U^{229}$  alpha decay is any different since their spacing and intensity variation is typical of that of other odd-mass nuclei in the heavy element region. This model will be used then for the interpretation of the  $Th^{225}$  energy levels.

From the Bohr-Mottelson model, the rotational energy for each member of band is given by:

$$E_I = \frac{\hbar^2}{2\mathcal{I}} [I(I+1) - I_0(I_0+1)] \quad (3)$$

where  $E_I$  is the energy above the ground state of the level of spin  $I$ , and



MU-23307

Fig. 13. Alpha decay scheme of  $U^{229}$ .

and  $\mathcal{I}$  is the moment of inertia.\* For odd mass nuclides, the allowable spins are  $I_0, I_0+1, I_0+2$ . For all members of a rotational band, the projection of the total angular momentum along the nuclear symmetry axis (K-quantum number) is constant and equal to  $I_0$ .

With the assumption that  $K = 3/2$  for this rotational band, and using the energy of the first excited state, the rotational constant  $\hbar^2/2\mathcal{I}$  is calculated to be 5.80 kev. Using this value in Equation 3 the energies of the higher members were calculated and are shown in Table II. It is seen that the agreement is very poor, the calculated values being too high. This is just the kind of effect expected for perturbations of the level spacings due to vibration-rotation interaction and centrifugal stretching. In order to take this effect into account a correction term of the form  $-B I^2 (I+1)^2$  was introduced into Equation 3 and the calculation repeated. These results are also shown in Table II. The agreement is seen to be much better for the  $9/2$  state. The rotational constant, 6.49 kev, is also in much better accord with the trend established by  $\text{Th}^{229}$  and  $\text{Th}^{231}$  of 6.2 and 5.9 kev respectively.

With the assumption that  $K \geq 5/2$ , fair agreement can be achieved for the energy level spacings but the rotational constant becomes smaller as K increases, a value of 4.4 kev being obtained for  $K = 5/2$ . Such a value does not seem at all reasonable. For  $K = 1/2$ , all the levels must be used to evaluate the constants. In this case the rotational constant is about 9 kev which seems too high but cannot be definitely ruled out since a value of 8.4 kev was obtained for the proposed  $K = 1/2$  band in  $\text{Ra}^{233.29}$ .

Since favored alpha decay takes place to this band, the theory of Bohr, Fröman, and Mottelson is therefore applicable to calculate the relative alpha intensities to the members of the band. The formula for calculating the relative transition probabilities is:

$$P = P_0(Z, E) \sum_L c_L \left[ \langle I_i L K_i K_f - K_i | I_i L I_f K_f \rangle + (-)^{I_f + K_f} b_L \langle I_i L K_i -K_f -K_i | I_i L I_f -K_f \rangle \right]^2 \quad (4)$$

\* In rotational spectra, for configurations with  $k = 1/2$ , an additional term  $E_I = \frac{\hbar^2}{2\mathcal{I}} a (-)^I + 1/2 (I + 1/2) \delta_{k, 1/2}$  must be added to Equation 3 where  $a$  is a decoupling parameter.

where  $P_0(Z, E)$  is the barrier penetration factor,  $c_L$  is the reciprocal of the hindrance factor for the alpha group of angular momentum  $L$  in neighboring even-even nuclei, the bracketed term is the Clebsch-Gordan coefficient describing the distribution of the alpha waves among various members of the rotational band, and the parameter  $b_L$  depends on the intrinsic wave function and arises from symmetrization of the wave function. (In the absence of detailed knowledge of the intrinsic wave function  $b_L$  is considered as a parameter to be evaluated from the experimental observations.)

In the calculation for decay from an  $I = K = 3/2$  state to a  $K = 3/2$  band the  $b_L$  term was ignored for two reasons: (a) since there was no appreciable alternation of intensities the  $b_L$  term in Equation 4 is small and (b) the coefficient  $\langle I_i L K_i - K_f - K_i | I_i L I_f - K_f \rangle$  vanishes for the cases where  $L < |K_i + K_f|$  and therefore affects only the components for which  $L \geq 4$ . The results of the calculation are shown in Table III where, upon comparison with theory, the agreement is found to be as good as that found for other nuclides studied.<sup>30</sup> The calculation was also performed assuming other values of  $K_i$  and  $K_f$  (for  $K_f = 1/2$  the  $b_L$  term was not ignored) but the agreement with experiment was found to be poor. In view of the foregoing arguments the ground state of  $U^{229}$  and  $Th^{225}$  are tentatively assigned a spin of  $3/2$ .

Besides determining the spin, it is of interest to determine the intrinsic state. For this purpose the Nilsson diagram for neutrons is employed (Figure 14). The ordinate represents not only an energy scale but also is used as a means of visualizing the order of filling the levels. The abscissa scale represents the deformation of prolate spheroid in terms of the parameter  $\delta$ . At zero deformation,  $\delta = 0$ , the levels are simply those of the shell model. As the deformation increases the  $(2j + 1)$ -fold degeneracy is reduced to 2-fold degeneracy and at high deformation the levels are described in terms of a new set of quantum numbers. The projection of the particle angular momentum,  $j$ , along the nuclear symmetry axis becomes a "good" quantum number; the number,  $\Omega$ , is the observed spin in the absence of rotational motion. The parity,  $\pi$ , is designated (+) or (-). The first number in the brackets,  $N$ , corresponds to the oscillator shell of the shell model. The second

Table II. Energies of  $\text{Th}^{225}$   $K = 3/2$  Rotational Band

| Spin of<br>State | Excited State Energy |                              |                                 |
|------------------|----------------------|------------------------------|---------------------------------|
|                  | Exp.                 | Theor. (Simple) <sup>a</sup> | Theor. (corrected) <sup>b</sup> |
| 3/2              | 0                    | (0)                          | (0)                             |
| 5/2              | 29                   | (29)                         | (29)                            |
| 7/2              | 65                   | 69                           | (65)                            |
| 9/2              | 102                  | 122                          | 103                             |

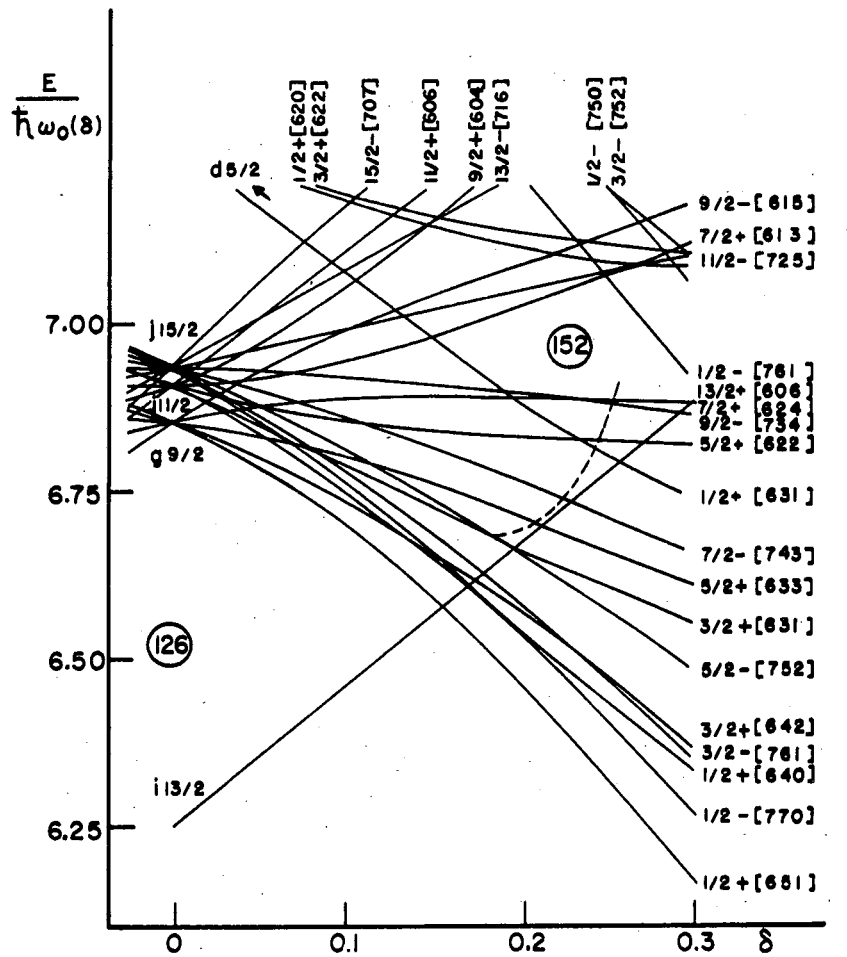
<sup>a</sup>  $\hbar^2/2\mathcal{I} = 5.80$  kev.

<sup>b</sup>  $\hbar^2/2\mathcal{I} = 6.49$  kev.  $B = 0.055$  kev.

Table III. Favored Alpha Intensities to  $\text{Th}^{225}$   $K = 3/2$  Band

| Excited-State<br>Energy (kev) | Spin | Relative Intensities |             |
|-------------------------------|------|----------------------|-------------|
|                               |      | Experimental         | Theoretical |
| 0                             | 3/2  | (64)                 | (64)        |
| 29                            | 5/2  | 20                   | 20          |
| 65                            | 7/2  | 11                   | 8.6         |
| 102                           | 9/2  | 1                    | 0.8         |

$C_0 = 1, C_2 = 1, C_4 = .091$



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Fig. 14. Nilsson diagram for neutrons in the region  $126 \leq N \leq 160$ . The dashed line is an approximate indication of the nuclear deformation.

number in the brackets is  $n_z$ , the number of nodal planes perpendicular to the symmetry axis; and the last number is  $\Lambda$ , the component of the orbital angular momentum along the symmetry axis. Stephens, et al.<sup>31</sup> have made a classification of the intrinsic states appearing in the heavy element region and have observed that qualitatively the agreement between the data and Nilsson diagrams is very good. The dashed line appearing in Figure 15 was taken from their paper and indicates roughly the deformations involved in the heavy element region. Their line ended below the  $5/2^+ (6,3,3)$  level but by extending the line, it crosses the  $5/2^- (7,5,2)$  level and then reaches the  $3/2^+ (6,3,1)$  for an occupation number  $N = 137$ .

It is seen that this is in agreement with the tentative spin assignment of  $3/2$  found for  $U^{229}$  and  $Th^{225}$ . The agreement observed by Stephens, et al. for other nuclides adds confidence to this assignment. The fact that both  $U^{229}$  and  $Th^{225}$  have the same ground state configurations implies that the state occupied by the odd particle in  $Th^{225}$  is vacated when the particle becomes paired.

It is also seen that at this point on the Nilsson diagram there is another close-lying level,  $5/2^- (7,5,2)$ . It is possible that the level observed at 139 kev is this state and the level 39 kev higher is the first excited rotational level of this intrinsic state. There is at present no supporting experimental evidence for this assignment.

## 2. Thorium-225

### a. Alpha Spectrum

After the alpha spectrum of  $Th^{225}$  and  $Ra^{221}$  was obtained (Figure 15, the high energy region of the spectrum of Figure 11a), identification of the peaks was necessary. In view of the half-life of  $Ra^{221}$ , and the amount of activity obtained, it was not feasible to prepare a  $Ra^{221}$  source using the same technique as that employed to obtain the  $Th^{225}$  sources mentioned earlier. A recoil collection was therefore tried inside the spectrograph, the collector mounted at the source position and the plate containing the activity placed in such a position that the alpha particles emitted by it would not strike the photographic plate. In order to degrade the energy of the recoils to permit collection with a reasonably low electric field, a film of zapon ( $5-10 \mu \text{ gm/cm}^2$ ) was placed over the activity. It was hoped that at least part of the  $U^{229}$

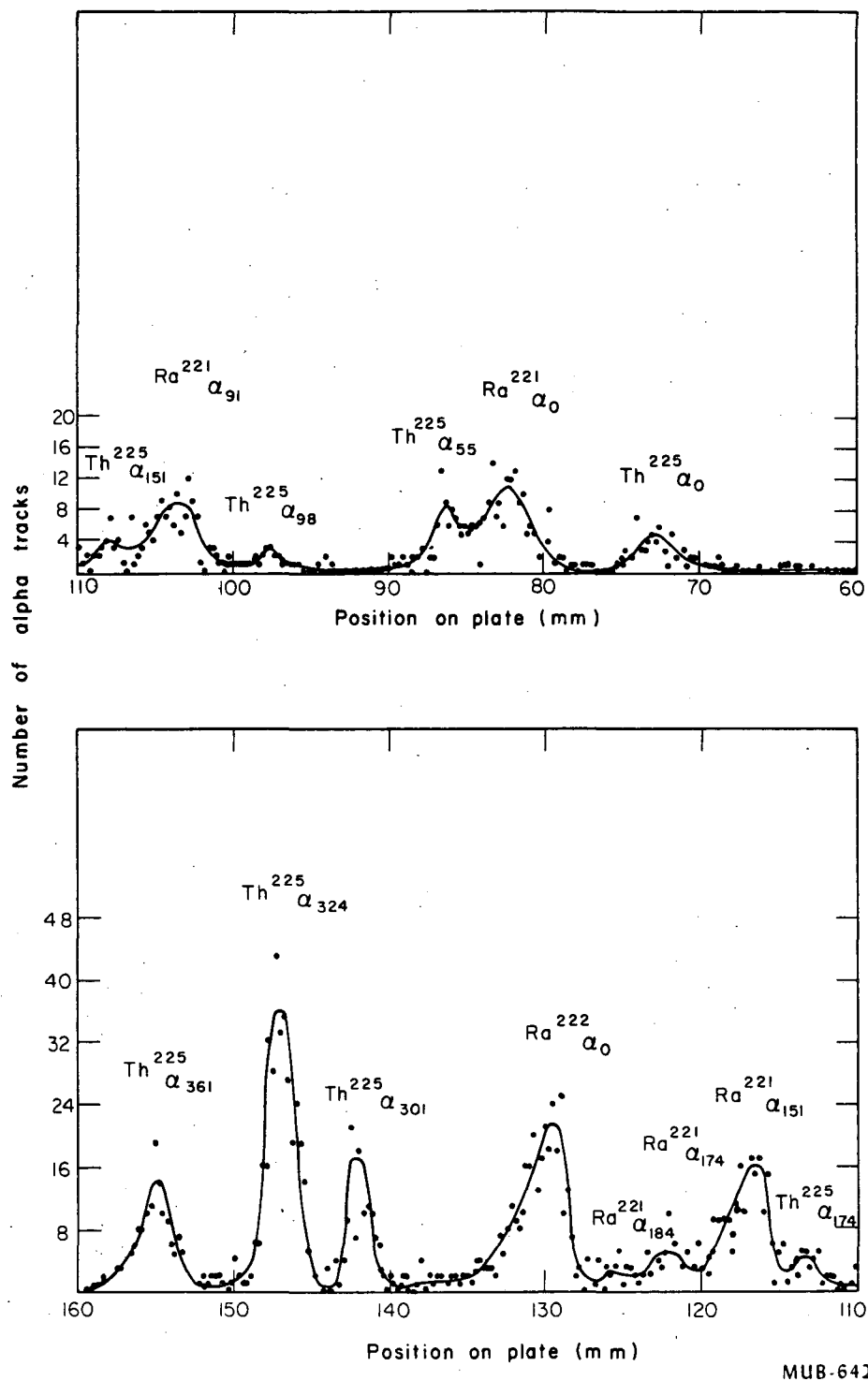


Fig. 15 Alpha spectrum of  $\text{Th}^{225}$  and  $\text{Ra}^{221}$  taken with alpha particle spectrograph.

recoils would be stopped with a higher percentage of the  $\text{Th}^{225}$  recoils getting through. Thus the source would be enriched in  $\text{Ra}^{221}$  which would increase the intensity of the  $\text{Ra}^{221}$  groups in the spectrum relative to those of  $\text{Th}^{225}$ . This technique failed because of the small number of recoils collected at the source position.

Somewhat indirect methods had to be relied upon for the identification. In a spectrum obtained with a  $\text{Th}^{225}$  source, the  $\text{Ra}^{221}$  peaks are broadened and exhibit low energy tailing because some of the  $\text{Ra}^{221}$  atoms are embedded in the source plate due to the recoil energy from  $\text{Th}^{225}$  decay. This effect is clearly shown (Figure 15) by a comparison of the  $\text{Th}^{225}\alpha_{324}$  and  $\text{Ra}^{222}\alpha_0$  groups.

Advantage was also taken of the  $\text{Ra}^{221}$  atoms lost from the source from  $\text{Th}^{225}$  decay. Since the recoil-collected sources were mass free this loss was appreciable--about 40 percent. On the other hand, the  $\text{U}^{229}$  sources always had some mass so their recoil loss was only about 5 percent. A comparison of the spectra obtained from  $\text{U}^{229}$  and  $\text{Th}^{225}$  sources thus revealed significant differences in intensity for  $\text{Ra}^{221}$  alpha groups relative to those of  $\text{Th}^{225}$ .

Both of these methods were used to make the alpha group assignments. The main limitation was the small number of alpha tracks obtained for the low intensity groups making their assignment somewhat less certain. Table IV summarizes the  $\text{Th}^{225}$  data obtained from an analysis of several spectra.

In the region of the  $\text{U}^{229}$  alpha groups this technique could not be used. Assignment of the groups  $\alpha_{460}$  and  $\alpha_{494}$  was made by comparing  $\text{Th}^{225}$  alpha spectra taken on the spectrograph with  $\text{Ra}^{221}$  alpha spectra taken with the alpha-particle grid chamber (Section C-1). The absence of the groups in the  $\text{Ra}^{221}$  spectrum identified them as  $\text{Th}^{225}$  groups.

#### b. Gamma-Alpha Coincidence Spectrum

The results of the gamma-alpha coincidence obtained in conjunction with  $\text{U}^{229}$  (Section 1-b) were also used for  $\text{Th}^{225}$ . A comparison of the Figures 12 shows a complete absence of the highest energy group in the coincidence spectrum compared to the singles spectrum. This indicates the group in 9 percent intensity designated as  $\alpha_0$  is indeed

Table IV. Alpha Groups of  $\text{Th}^{225}$

| Alpha-Particle<br>Energy (Mev) <sup>a</sup> | Abundance<br>% | Excited-State<br>Energy (kev) <sup>b</sup> | Hindrance<br>Factor <sup>c</sup> |
|---------------------------------------------|----------------|--------------------------------------------|----------------------------------|
| 6.793 $\pm$ 0.005                           | 9 $\pm$ 1      | 0                                          | 1.8 $\times 10^2$                |
| 6.739                                       | 7 $\pm$ 1      | 55                                         | 1.4 $\times 10^2$                |
| 6.695                                       | 2 $\pm$ 1      | 98                                         | 3.4 $\times 10^2$                |
| 6.645                                       | 3 $\pm$ 1      | 151                                        | 1.4 $\times 10^2$                |
| 6.622                                       | 3 $\pm$ 1      | 174                                        | 1.1 $\times 10^2$                |
| 6.496                                       | 14 $\pm$ 1     | 301                                        | 7.3                              |
| 6.473                                       | 43 $\pm$ 2     | 324                                        | 1.9                              |
| 6.436                                       | 15 $\pm$ 1     | 361                                        | 3.8                              |
| 6.340                                       | 2 $\pm$ 1      | 460                                        | 11                               |
| 6.307                                       | 2 $\pm$ 1      | 494                                        | 7.7                              |

<sup>a</sup> Alpha particle energies are expressed relative to  $\text{Th}^{226}$   $\alpha_0$  and  $\text{Ra}^{222}$   $\alpha_0$  taken as 6.330 and 6.552 Mev respectively.

<sup>b</sup> The limits of error are  $\pm 5$  kev for all groups except for the three most intense groups where they are  $\pm 3$  kev.

<sup>c</sup> The hindrance factors were calculated using an alpha branching ratio for  $\text{Th}^{225}$  of 0.95. A quantitative comparison of the  $\text{U}^{229}$  and  $\text{Th}^{225}$  alpha peaks revealed that the alpha branching ratio for  $\text{Th}^{225}$  was  $\geq 0.95$ , and not 0.9 as previously reported.

the ground state transition or is within the L-electron binding energy of the ground state group.

c. Gamma Spectra

(1) Total Alpha-Gamma Coincidence Spectra:

The alpha-gamma coincidence spectrum of the  $U^{229}$  family is shown in Figure 16. The dotted line shows the contribution from the  $Ra^{221}$  gamma rays (obtained by running an alpha-gamma coincidence spectrum on the second order recoils of  $U^{229}$ ). Figure 17 shows the high energy gamma ray spectrum. Table V shows the results of this study. These data have been corrected for the  $Th^{226}$  family contribution; its contribution was determined by repeating the experiment after the  $U^{229}$  had decayed.

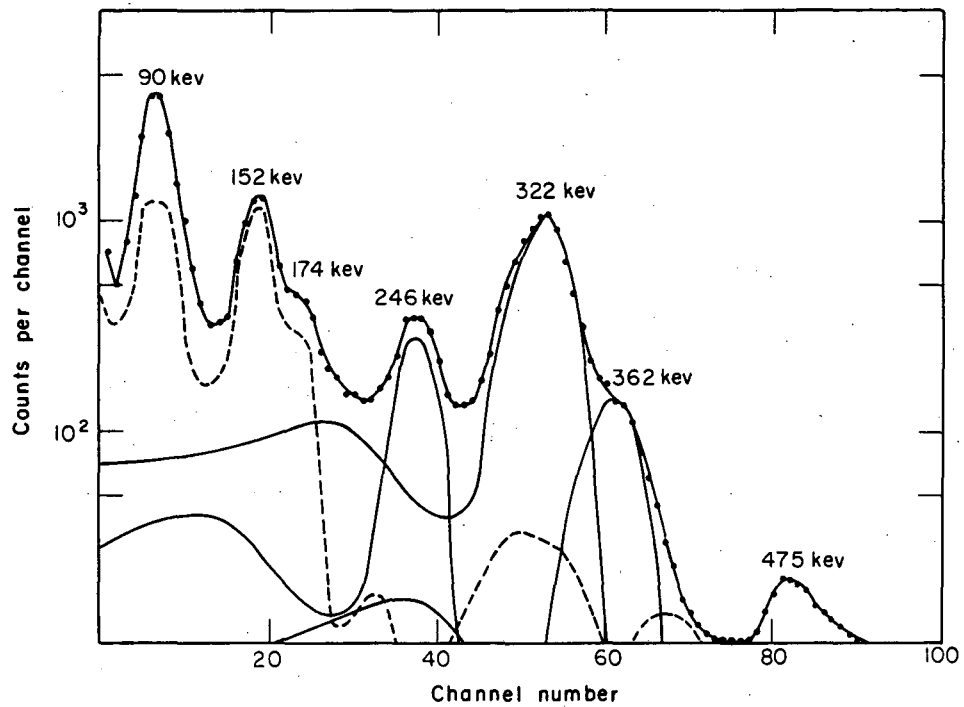
(2) Specific Alpha-Gamma Coincidence Spectra:

Fortunately the three most intense groups of  $Th^{225}$  could be gated on with essentially no interference from the  $Ra^{221}$  in equilibrium with it. However, its presence prevented a study of the lower intensity alpha groups, since the high intensity  $Ra^{221}$  groups occurred at about this same energy (Section 3a). The spectra obtained by gating on  $\alpha_{301}$ ,  $\alpha_{324}$ , and  $\alpha_{361}$ , are shown in Figures 18-20. The data are summarized in Table VI.

In these data, the relative intensities are more significant than the absolute intensities. It is obvious that the 246, 322, and 362 keV gamma rays are in coincidence with  $\alpha_{301}$ ,  $\alpha_{324}$ , and  $\alpha_{361}$ , respectively. Since the alpha groups were not completely resolved by the grid chamber, the alpha group nearest the gating group was partly included in the gate. This explains the appearance of the 322 and 362 keV gammas when gating on the  $\alpha_{301}$  and  $\alpha_{322}$ , respectively.

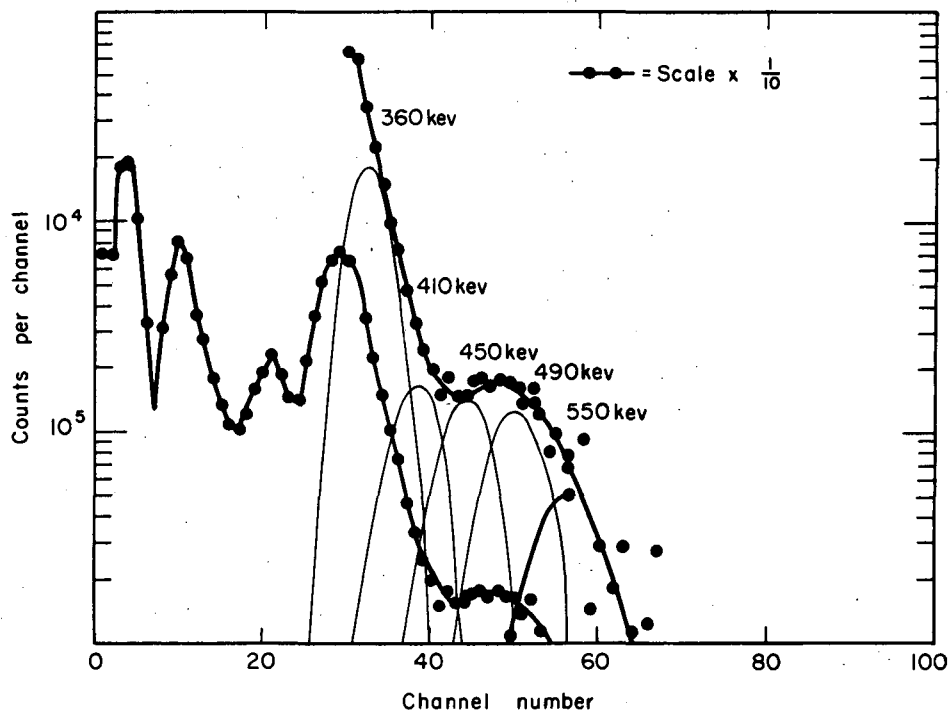
TABLE V

| Gamma Rays of $Th^{225}$ |                         |
|--------------------------|-------------------------|
| Energy (keV)             | Intensity Photons/Alpha |
| $90 \pm 2$               | $0.33 \pm 0.03$         |
| $246 \pm 2$              | $0.05 \pm 0.01$         |
| $322 \pm 2$              | $0.30 \pm 0.03$         |
| $362 \pm 3$              | $0.05 \pm 0.01$         |
| 450                      | 0.01                    |
| 490                      | 0.01                    |



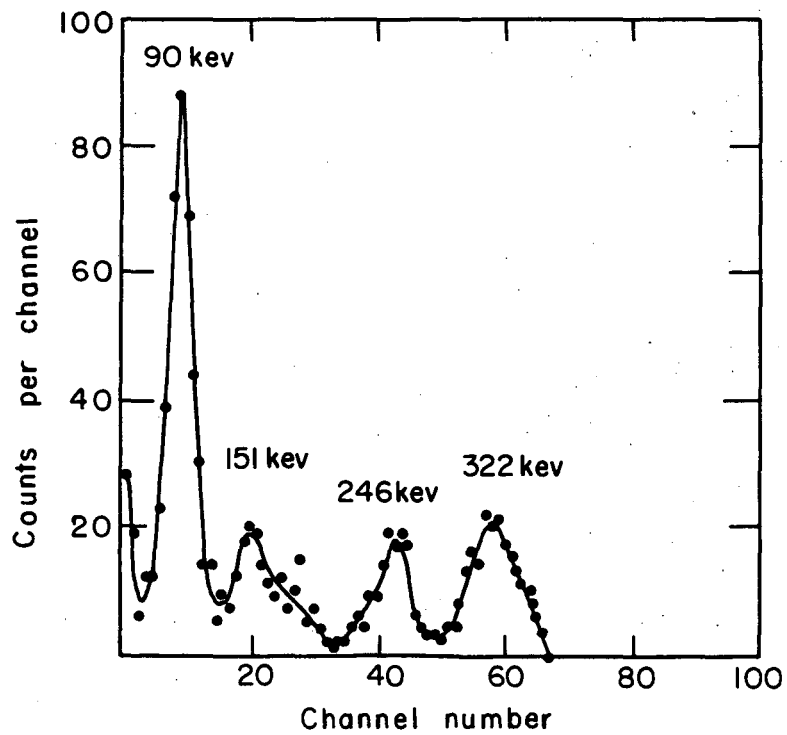
MU-23309

Fig. 16. Alpha particle - gamma ray coincidence spectrum of  $\text{Th}^{225}$  and  $\text{Ra}^{221}$  (low energy).



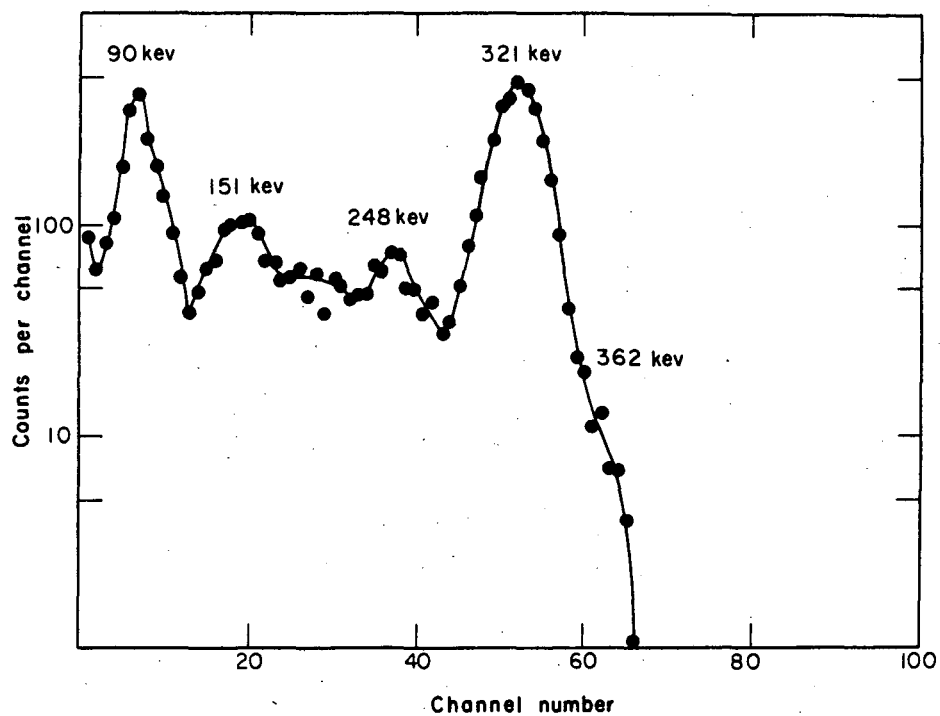
MU-23310

Fig. 17. Alpha particle - gamma ray coincidence spectrum of  $\text{Th}^{225}$  and  $\text{Ra}^{221}$ .  
(high energy).



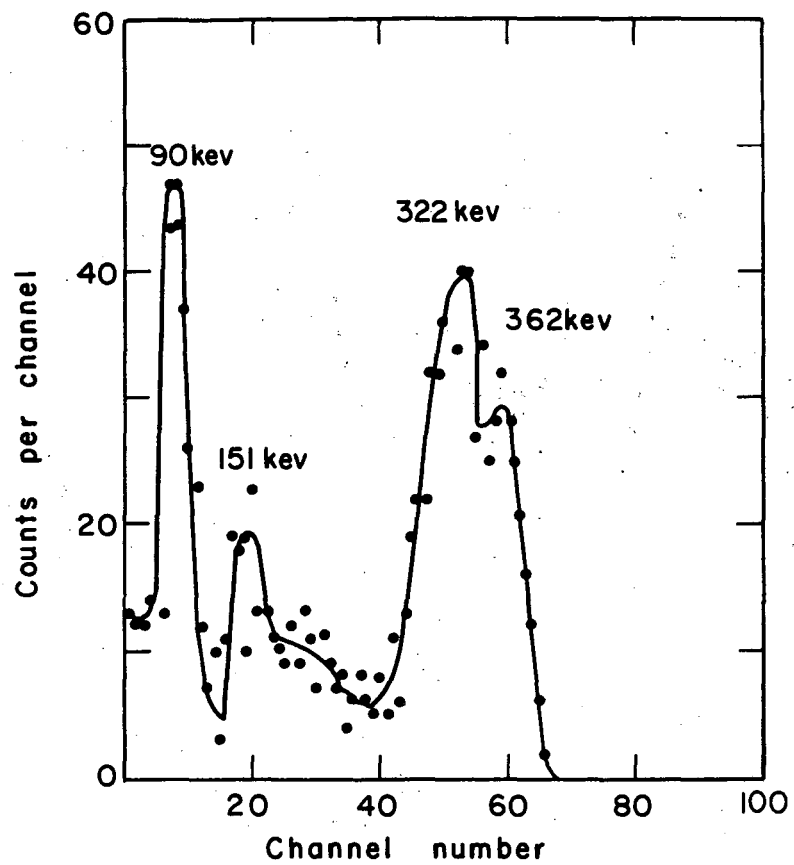
MU-23311

Fig. 18.  $\text{Th}^{225}\alpha_{301}$  - gamma ray coincidence spectrum.



MU-23312

Fig. 19.  $\text{Th}^{225} \alpha_{324}$  - gamma ray coincidence spectrum.



MU-23313

Fig. 20.  $\text{Th}^{225}\alpha_{361}$  - gamma ray coincidence spectrum.

Table VI. Summary of Specific Alpha-Gamma Coincidence Study

| Alpha<br>Group | Coincident Radiation<br>(kev) | Intensity - Photons/Alpha |           |
|----------------|-------------------------------|---------------------------|-----------|
|                |                               | Uncorrected               | Corrected |
| $\alpha_{301}$ | 90                            | 0.12                      | 0.08      |
|                | 135                           | 0.01                      | 0.004     |
|                | 246                           | 0.03                      | 0.03      |
|                | 322                           | 0.06                      | 0         |
| $\alpha_{322}$ | 90                            | 0.11                      | 0.06      |
|                | 151                           | 0.01                      | 0.01      |
|                | 248                           | 0.02                      | 0         |
|                | 322                           | 0.30                      | 0.30      |
|                | 362                           | 0.005                     | 0         |
| $\alpha_{361}$ | 90                            | 0.03                      | 0.01      |
|                | 150                           | 0.01                      | 0.004     |
|                | 305                           | 0.01                      | 0.01      |
|                | 322                           | 0.06                      | 0         |
|                | 362                           | 0.05                      | 0.05      |

When the lower energy gamma ray appeared as a "contaminant" in the spectrum, it was not possible to tell whether it was due to gating on part of another alpha group or whether the level populated by the gating alpha group de-excited to this level causing the lower energy gamma ray; an example is the appearance of the 246 kev gamma when gating on  $\alpha_{324}$ . The contribution of gamma rays to the coincidence spectra arising from either of these causes has been subtracted and the corrected intensities appear in the last column of Table VI.

Some inferences concerning the multipolarities of these gamma rays can be made using the data of Table VI, and the intensities of the alpha groups to the states giving rise to the transitions. For the purposes of the analyses it is assumed that the 90 kev radiation observed in Table VI is due to K x-rays. (It will be seen that this assumption affects only the arguments concerning the 246 kev transition.) The intensity of K x-rays associated with the 246 kev transition suggests that it is magnetic. The total intensity of such a transition could be accounted for by the alpha population to the 301 kev level.

From the K x-ray intensity appearing in the spectrum with the 322 kev gamma ray, the transition must be electric. It is not possible to tell whether it is E1 or E2 since the transition intensity for either case would not exceed the alpha population to the state. An E2 transition can account for 34 of the 43 percent population from the state. Therefore, the level must also de-excite by other modes which accounts for the 151 kev radiation in coincidence with  $\alpha_{322}$ .

The K x-ray intensity in coincidence with  $\alpha_{361}$  suggests that this transition is also electric with the choices being E1 or E2. It cannot be said whether or not the 361 level de-excites to the level at 322 kev because even though the 322 kev gamma appears in the same spectrum it may be due to the incomplete resolution of the gating alpha groups. It is certain, however, that very little if any de-excitations occur to the level at 301 kev. The 305 kev gamma ray is possibly the transition to the level at 55 kev.

The energies of the 450 and 490 kev gamma rays compare reasonably well with the energy levels at 460 and 494 kev. The gamma rays are tentatively indicated as de-exciting these levels to the ground state.

d. Decay Scheme

A decay scheme summarizing the alpha decay data on  $\text{Th}^{225}$  is presented in Figure 21. A solid line indicates transitions that are certain while a broken line indicates proposed transitions.

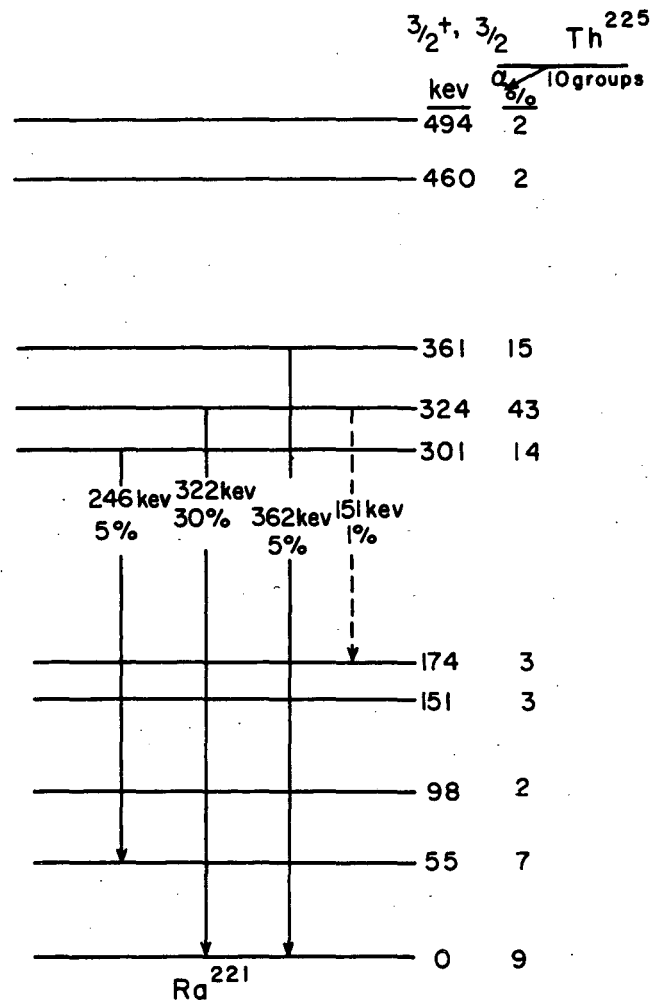
e. Interpretation

In considering the decay scheme of  $\text{Th}^{225}$ , attention is drawn to the highly populated level at 324 kev. Since the alpha hindrance factor is close to 1 (Table V) then in terms of the Bohr-Mottelson theory this can be interpreted as favored alpha decay from the ground state of  $\text{Th}^{225}$ . Since this has been assigned a spin of  $3/2$  then the 324 kev level in  $\text{Ra}^{221}$  should be  $I = K = 3/2$ . Therefore rotational levels would be expected to appear at higher energies. The level at 361 kev is the only possibility for the first excited rotational state. The energy difference is 37 kev from which the value of the rotational constant,  $\hbar^2/2\mathcal{I}$  is calculated to be 7.4 kev. Since no correction term could be included in the formula for rotational energy level spacing, this value is probably low. From a consideration of the value of the rotational constants at slightly higher mass numbers, this value might be expected to be about 10 kev. Thus another level is required in the region 80 to 100 kev above the base level. Its intensity (calculated using Equation 4) should be about 3 to 4 percent. No state was observed in this energy region. The closest group lies at 136 kev above the base rotational level which cannot be interpreted as a rotational member in terms of this model. Further discussion will be presented in Section 6.

3. Radium-221

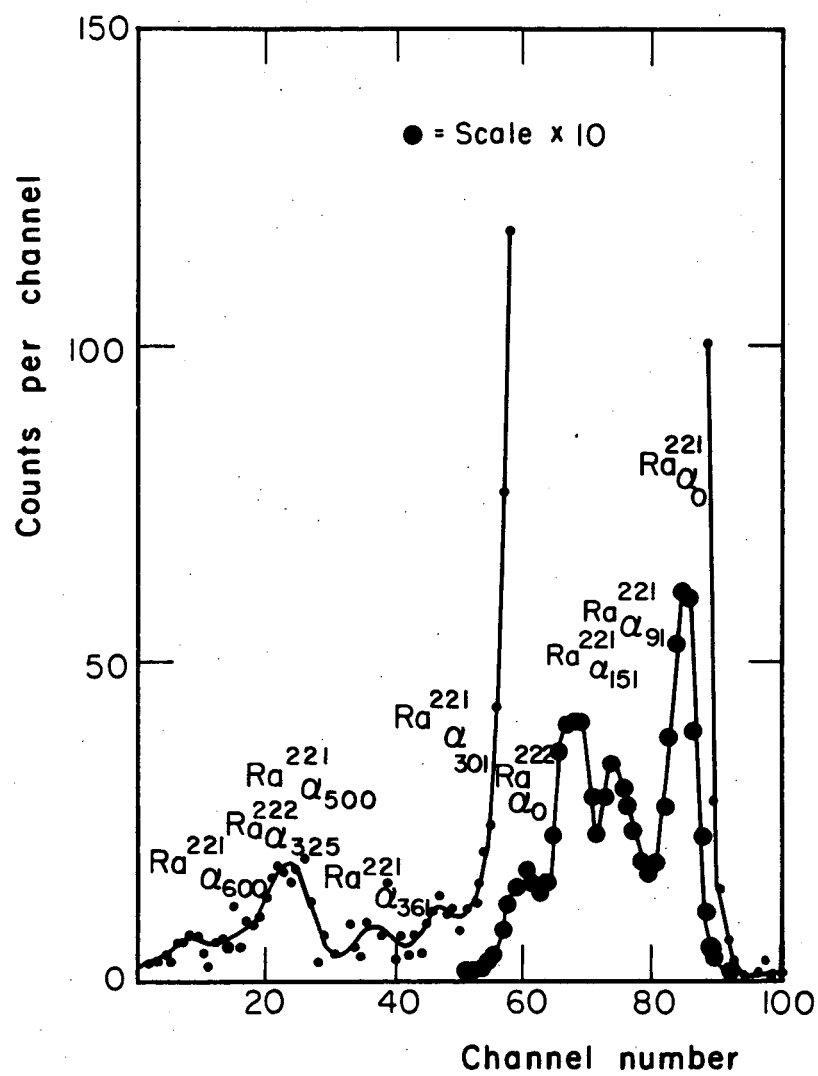
a. Alpha Spectrum

The method used in obtaining the alpha spectrum on the spectrograph has already been discussed in connection with  $\text{Th}^{225}$  (Section 2a) and Figure 15. The  $\text{Ra}^{221}$  alpha spectrum free from  $\text{Th}^{225}$  was also obtained using the alpha particle grid chamber. The source was prepared by performing a double recoil collection from the  $\text{U}^{229}$  grandparent using collection times and counting times of one minute. A typical spectrum obtained in a 2-hour run is shown in Figure 22. The existence of the three most intense groups ( $\alpha_{174}$  and  $\alpha_{184}$  are not resolved from  $\alpha_{151}$ ) is confirmed. The group designated as the  $\alpha_0$  exhibits an increased



MU-23314

Fig. 21. Alpha decay scheme of Th  $^{225}$ .



MU-23315

Fig. 22 Alpha spectrum of  $\text{Ra}^{221}$  taken with alpha particle grid chamber.

intensity because the conversion electrons from transitions to this state have not been collimated out. The intensities listed in Table VIII are those taken from the spectrograph data.

Besides these groups, the existence of four low intensity groups is indicated. The group designated as  $\alpha_{500}$  is clearly in evidence. Although the other three groups were observed in two other spectra, the small number of events renders their existence somewhat less certain. These groups have been indicated by parentheses in Table VII where the alpha decay data are tabulated.

TABLE VII

| Alpha Groups of Ra <sup>221</sup>        |             |                                         |                     |
|------------------------------------------|-------------|-----------------------------------------|---------------------|
| Alpha-particle energy (Mev) <sup>a</sup> | Abundance % | Excited state energy (Mev) <sup>b</sup> | Hindrance factor    |
| 6.754 ± 0.005                            | 30 ± 2      | 0                                       | 14                  |
| 6.661                                    | 20 ± 2      | 0.091                                   | 10                  |
| 6.606                                    | 34 ± 2      | 0.151                                   | 3.0                 |
| 6.584                                    | 8 ± 1       | 0.174                                   | 11                  |
| 6.574                                    | 3 ± 1       | 0.184                                   | 28                  |
| (6.46)                                   | 0.4 ± 0.3   | 0.30                                    | 7 × 10 <sup>1</sup> |
| (6.40)                                   | 0.3 ± 0.2   | 0.36                                    | 5 × 10 <sup>1</sup> |
| 6.25                                     | 0.7 ± 0.3   | 0.50                                    | 6                   |
| (6.16)                                   | 0.3 ± 0.2   | 0.60                                    | 5                   |

<sup>a</sup> Alpha particle energies are expressed relative to Th<sup>226</sup>,  $\alpha_0$  and Ra<sup>222</sup> $\alpha_0$  taken as 6.330 and 6.552 Mev, respectively.

<sup>b</sup> For groups having intensities less than 1 percent (those determined with the grid chamber) the limits of error are ± 10 kev; and ± 3 kev for the rest.

TABLE VIII

| Gamma Rays of Ra <sup>221</sup> |                           |
|---------------------------------|---------------------------|
| Energy (kev)                    | Intensity (Photons/Alpha) |
| 89 ± 2                          | 0.15 ± 0.02               |
| 152 ± 2                         | 0.13 ± 0.02               |
| 176 ± 2                         | 0.02 ± 0.005              |
| 219 ± 10                        | 0.001                     |
| 293                             | 0.006                     |
| 320                             | 0.007                     |
| 415 ± 10                        | 0.005                     |

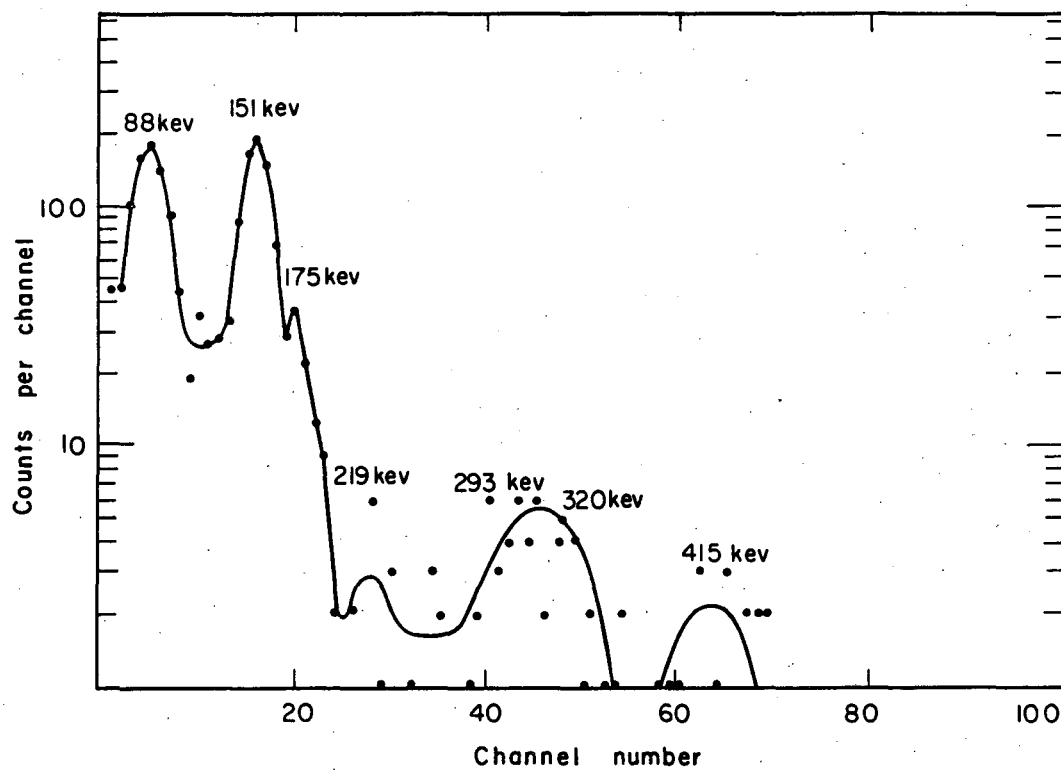
b. Gamma-Alpha Coincidence Spectrum

Reference is again made to Figure 12b in which the coincidence spectrum of Ra<sup>221</sup> was also obtained. A comparison with Figure 12a reveals that the group at 6.754 Mev is within the L-electron binding energy of the ground state.

c. Gamma Spectra

(1) Total Alpha-Gamma Ray Coincidence Spectrum:

This source was prepared by a double recoil collection from U<sup>229</sup>. The collection time was one minute for the Ra<sup>221</sup> and the counting time was one minute. By exchanging sources every minute a total of 2 hours counting time was recorded. After the U<sup>229</sup> had decayed, the operation was repeated in order to correct for the Ra<sup>222</sup> contribution, which was small. The corrected spectrum is shown in Figure 23 and the data presented in Table VIII.



MU-23316

Fig 23. Alpha particle - gamma ray coincidence spectrum of  $\text{Ra}^{221}$ .

(2) Specific Alpha-Gamma Ray Coincidence Studies:

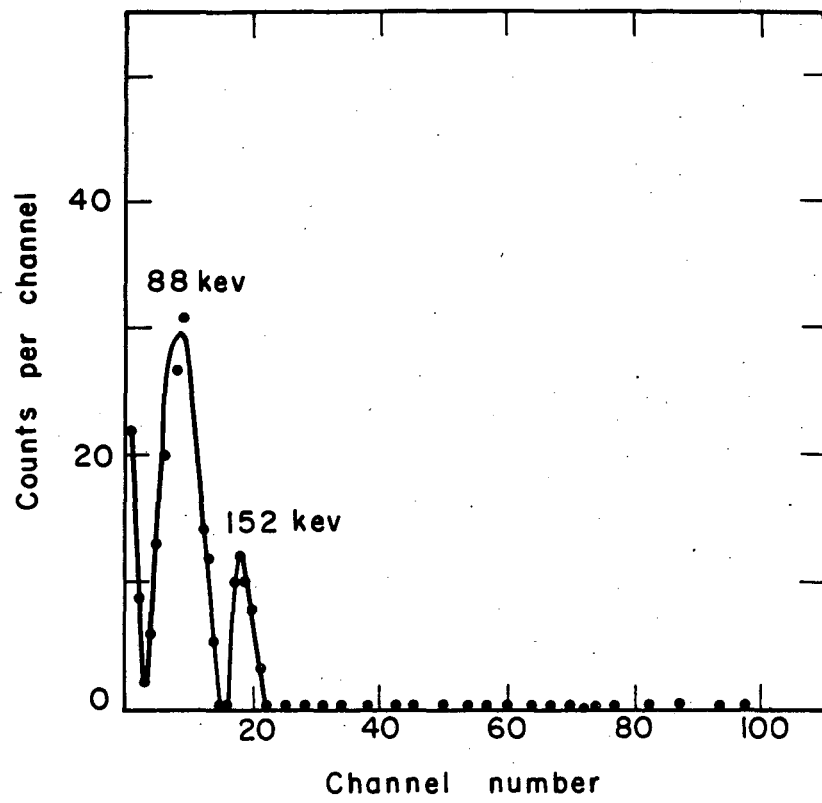
Three Ra<sup>221</sup> alpha groups were used as gates:  $\alpha_{91}$ ,  $\alpha_{151}$ , and  $\alpha_{174}$ . This was done using U<sup>229</sup> as the source. It readily can be seen that the intense groups of U<sup>229</sup> and Th<sup>225</sup> do not lie in this energy region (Table I and IV) so that there is essentially no interference from them. However, the alpha groups from these other nuclides made it impossible to study the de-excitation of the low intensity groups. The spectra obtained are shown in Figures 24-26 and the data are summarized in Table IX. The data were treated in the same way as that of Th<sup>225</sup> (Section 2c-2). These data definitely show that the 91, 151, and 176 kev gamma rays are in coincidence with  $\alpha_{91}$ ,  $\alpha_{151}$ , and  $\alpha_{174}$ , respectively and so must de-excite the corresponding levels to the ground state.

TABLE IX

| Summary of Specific Alpha-Gamma Coincidence Study |                            |                         |                         |
|---------------------------------------------------|----------------------------|-------------------------|-------------------------|
| Alpha group                                       | Coincident Radiation (kev) | Intensity — Uncorrected | Photons/Alpha Corrected |
| 91                                                | 91                         | 0.038                   | 0.035                   |
|                                                   | 151                        | 0.004                   | 0                       |
| 151                                               | 91                         | 0.11                    | 0.11                    |
|                                                   | 151                        | 0.10                    | 0.10                    |
|                                                   | 176                        | 0.01                    | 0                       |
| 174                                               | 90                         | 0.11                    | 0.01                    |
|                                                   | 151                        | 0.10                    | 0                       |
|                                                   | 176                        | 0.03                    | 0.03                    |

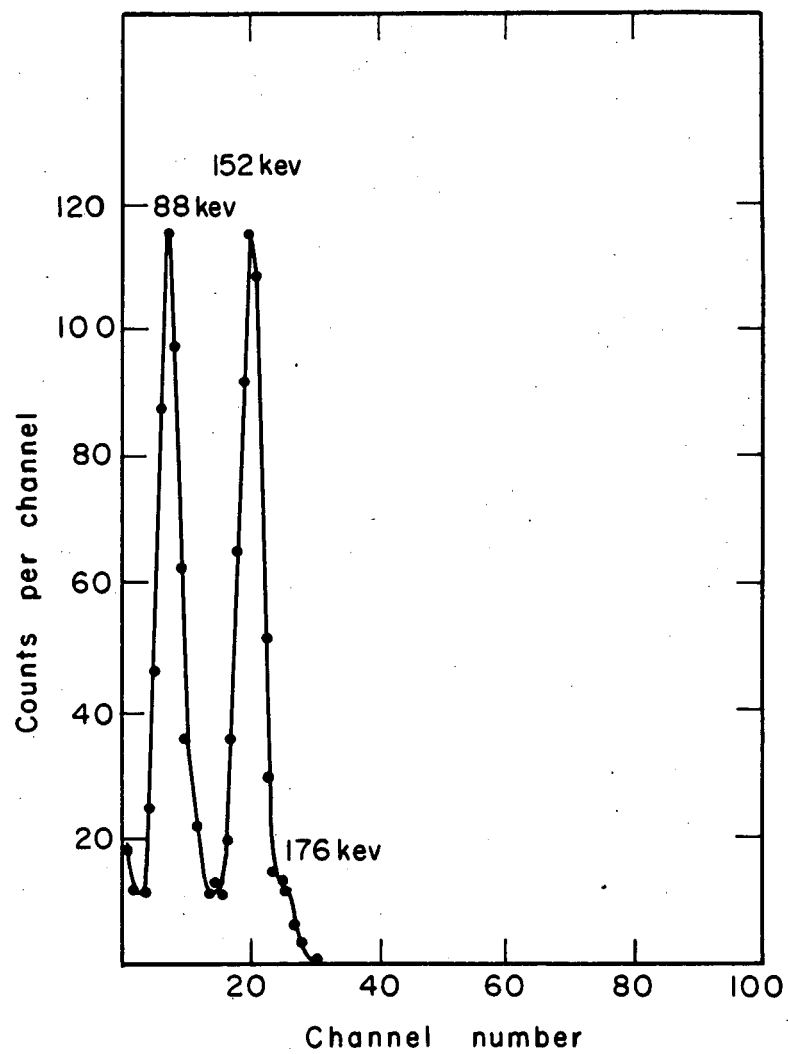
In the corrected  $\alpha_{91}$ -gamma coincidence results, the 91 kev gamma-ray has an intensity of 3.5 percent. Since the alpha population to the state is 20 percent, then the multipolarity of the transition must be either E1 or M1. If the assumption is made that there are no levels between the 90 kev level and ground populated by the decay of this state, then the transition must clearly be M1 with a conversion coefficient of 5 compared with that for an E1 of 0.2.

Attention will now be directed to the multipolarity of the 152 kev transition. A consideration of the de-excitation of all levels above the 151 kev level to this state yields a maximum population of 43 percent



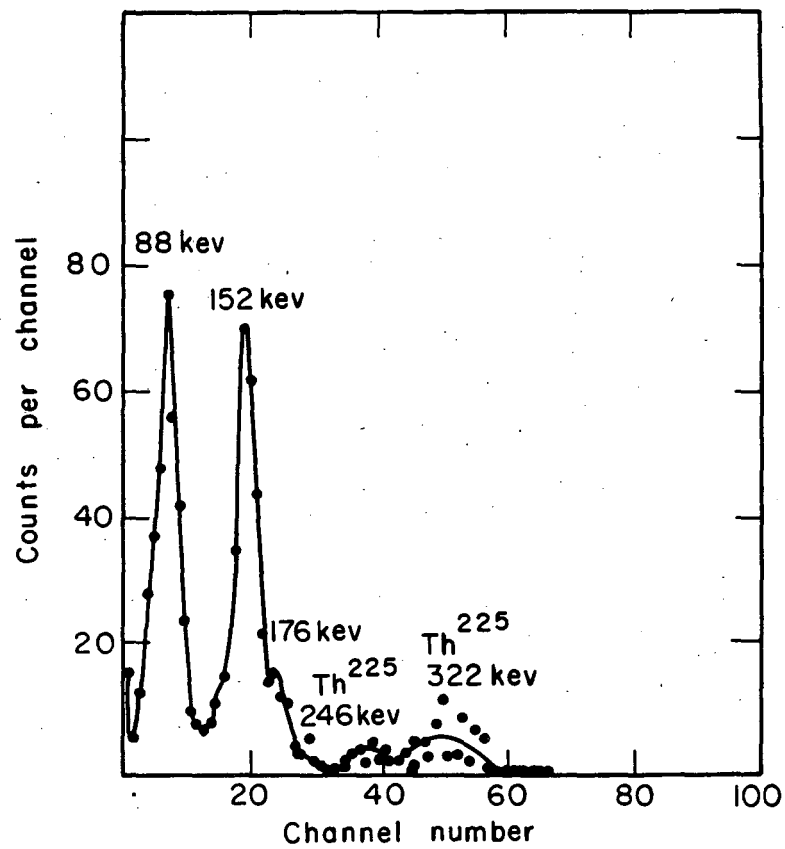
MU-23317

Fig 24.  $\text{Ra}^{221}_{91}$  - gamma ray coincidence spectrum.



MU-23318

Fig. 25.  $\text{Ra}^{221}\alpha_{151}$  - gamma ray coincidence spectrum.



MU - 23319

Fig. 26.  $\text{Ra}^{221}\alpha_{174}$  - gamma ray coincidence spectrum.

for this level; the minimum is 34 percent (the alpha intensity to the state). Since the de-exciting gamma ray has an intensity of 13 percent, the residual population of the state is between 21 and 30 percent. The transition cannot be purely electric because of the relatively high K x-ray intensity; an E2 transition would account for only 4 percent of the K x-ray intensity. An alternative source of 91 kev radiation could come from the 151 kev level de-exciting to the 91 kev level. However, this mechanism could account for a maximum of 5 percent. Clearly then the transition must have a magnetic admixture (it cannot be all magnetic because the population to the state is not great enough, i.e., about 80 percent population would be needed for an M1 transition). The K x-ray intensity could be explained by either an E2 or an M2-E1 mixture. From the data it is not possible to distinguish between these two choices.

The K x-ray intensity associated with the de-excitation of the 174 kev level indicates that the transition is predominately electric, either E1 or E2. Since the alpha population to the state is 8 percent, an E2 transition appears to be the most likely possibility.

Nothing can be said about the multipolarity of the other transitions.

#### d. Decay Scheme and Interpretation

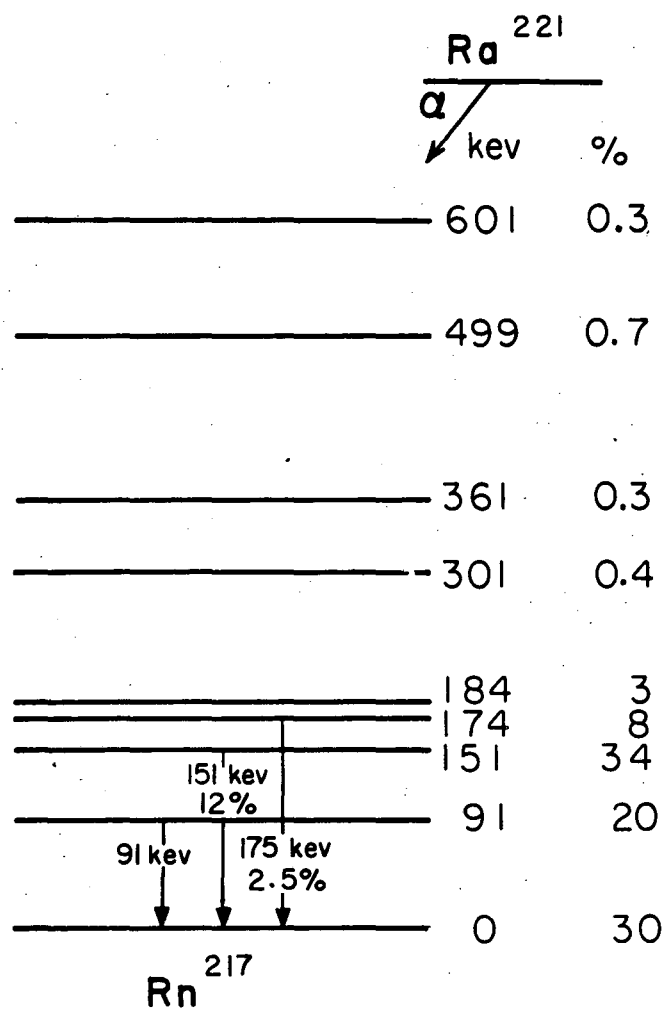
The decay scheme of  $\text{Ra}^{221}$  is shown in Figure 27. Only those transitions that are considered certain are shown. It is readily seen that on the basis of this data, a unique choice cannot be made for the placement of the other gamma rays in the decay scheme and so they have not been included.

No interpretation of the decay scheme can be given but a general discussion is presented in Section 6.

### 4. Radon-217

#### a. Alpha Spectrum

Because of the relatively high energy of  $\text{Rn}^{217}$  alpha particles, two different field settings (and thus two runs) had to be used to span the region from  $\text{Th}^{225}\alpha_0$  to  $\text{Rn}^{217}\alpha_0$ . In the energy region from 6.78 ( $\text{Th}^{225}\alpha_0$ ) to 7.680 Mev ( $\text{Po}^{214}$ ) only one alpha group was observed at an energy of 7.13 Mev which was due to  $\text{Rn}^{218}$ . The limit of detection was 1 percent.



MU-23320

Fig. 27. Alpha decay scheme of  $\text{Ra}^{221}$ .

A group at an energy of  $7.735 \pm 0.004$  Mev (calculated relative to the  $\text{Po}^{214}$  alpha group taken as 7.680 Mev) was observed to decay with the half-life of the  $\text{U}^{229}$  parent (1 hour), and was assigned to  $\text{Rn}^{217}$ . This region of the spectrum is shown in Figure 28. The hindrance factor was calculated to be 1.59 using the half-life of  $5.4 \times 10^{-4}$  sec (Section c).

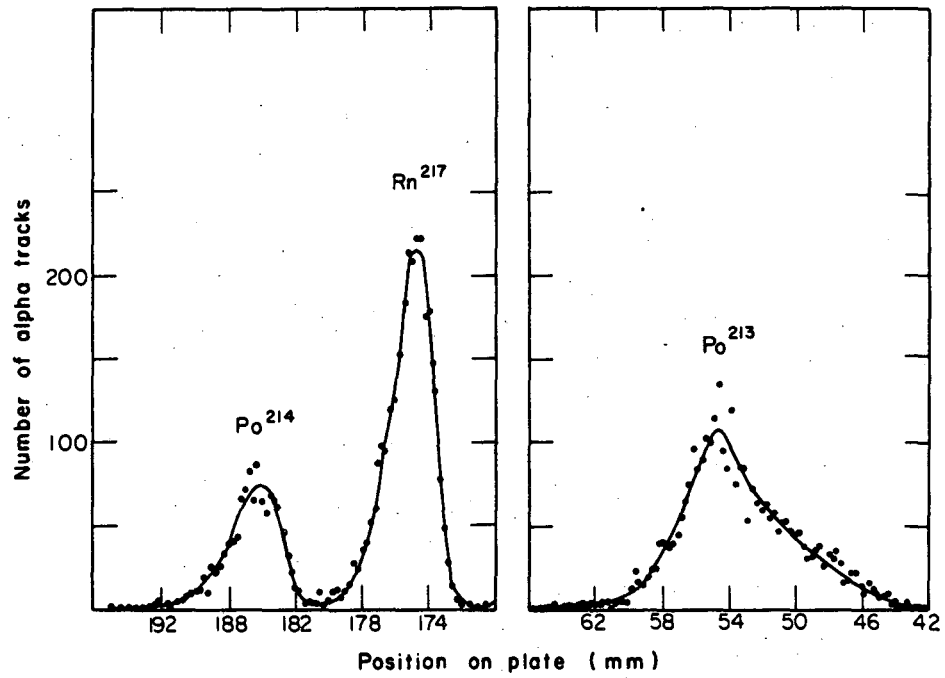
#### b. Gamma-Alpha Coincidence Spectrum

In order to extend the limit of detection and also to ascertain whether or not the 7.735 Mev alpha group populated the ground state in  $\text{Po}^{213}$ , a gamma-alpha coincidence experiment was run. This extended the limit of observation to 0.1 percent.

The 7.735 Mev alpha group was not found in coincidence indicating the daughter state to be within the L-electron binding energy of the ground state. There was some evidence for a group at 7.50 Mev in an intensity of about 0.1 percent. If the group exists, it could belong either to  $\text{Rn}^{217}$  or  $\text{Po}^{213}$ . If it is associated with the decay of  $\text{Po}^{213}$ , then a state must exist about 900 kev above the ground state. A state has been seen at 750 kev in  $\text{Pb}^{209}$  by the (d,p) reaction on  $\text{Pb}^{208}$ . If this were the state excited in the alpha decay the energies should be in better agreement. If this is not the same state, then a state at 900 kev might be expected to have been seen in the nuclear reaction study. However, its absence does not conclusively indicate that there is no level at this energy since selection rules may have prevented a very substantial population of the state. If the group is associated with the decay of  $\text{Rn}^{217}$ , then an excited state exists at 0.23 Mev. It is not possible to assign the group on the basis of the data.

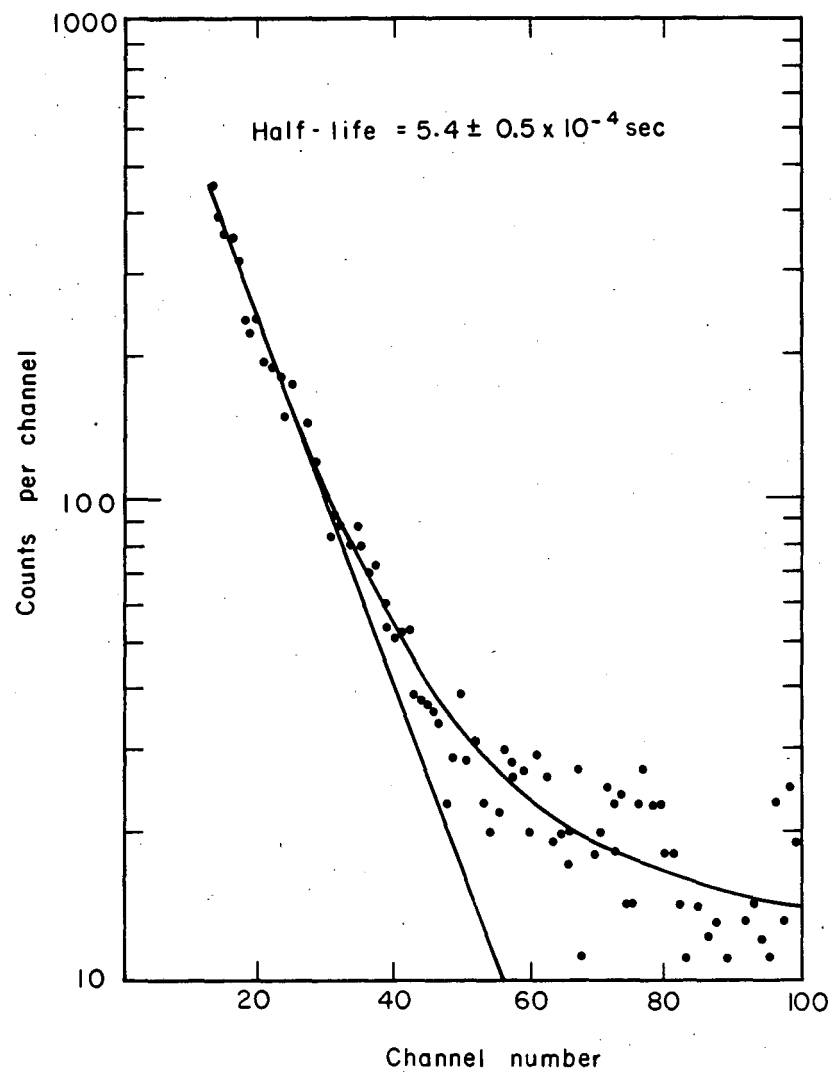
#### c. Half-Life Determination

The half-life was determined using the electronic system outlined in Section II, C-1. The  $\text{Ra}^{221}\alpha_0$  group supplied the trigger pulses and the  $\text{Rn}^{217}\alpha_0$  group supplied the signal pulses. Sweep times of 5 and 10 milliseconds were used with count rates of a few hundred counts per minute. A typical decay curve is shown in Figure 29, obtained with a sweep time of 10 milliseconds. The half-life obtained from five determinations was  $5.4 \pm 0.5 \times 10^{-4}$  seconds. This is a factor of two shorter than the previously reported value of  $10^{-3}$  seconds.<sup>23</sup>



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Fig. 28. Alpha spectrum of  $Rn^{217}$  and  $Po^{213}$  taken with alpha particle spectrograph.



MU-23352

Fig. 29. Delay coincidence curve of  $\text{Rn}^{217}$ .

d. Decay Scheme and Interpretation

The levels of  $\text{Po}^{213}$  have been studied from the beta decay of  $\text{Bi}^{213}$ . Wagner et al.<sup>34</sup> found two components in the beta spectrum: 1.39 (68 percent) and 0.959 Mev (32 percent). A gamma ray of 434 kev was also observed in agreement with the energy differences in the beta spectrum. Stephens<sup>35</sup> found from the number of K x-rays associated with the transition that the K conversion coefficient was 0.2, in good agreement with that expected for an M1 transition.

No alpha transition was seen which could populate the 434 kev level in  $\text{Po}^{213}$  with a limit of 0.05 percent indicating a hindrance factor of greater than 140. No level assignments can be made on the basis of the data (or theory).

5. Polonium-213

Alpha Spectrum and Gamma-Alpha Coincidence Spectrum

The alpha spectrum of  $\text{Po}^{213}$  was obtained in the same exposure as that for  $\text{Rn}^{217}$  (Figure 28). Because of the great difference in energy between the alpha group of  $\text{Po}^{213}$  and that of the standard,  $\text{Po}^{214}$ , they had to be focused on opposite ends of the plate. This is the reason for the high energy tailing on the  $\text{Po}^{213}$  peak. Its energy was determined to be  $8.37 \pm 0.10$  Mev. No other alpha groups were observed with the limit being 1 percent. The energy is in agreement with that obtained on the alpha particle grid chamber in conjunction with the  $\text{U}^{228}$  family experiments (Section B). However, it is higher by 0.02 Mev than the previously reported value of  $8.351 \pm 0.010$  Mev.<sup>35</sup>

In the coincidence spectrum of  $\text{Em}^{217}$  the spectrum of  $\text{Po}^{213}$  was also recorded. The absence of the ground state alpha group indicated that this group populated a level within the L-electron binding energy of the ground state. This level has been identified as  $g\ 9/2$ .<sup>30</sup>

6. Discussion

The nuclides in the  $\text{U}^{229}$  series span the region of transition between nuclei having a permanent spheroidal deformation and those being spherical. The spectra observed clearly reflect this fact. In  $\text{Th}^{225}$  rotational levels are observed characteristic of the strong coupling region. In  $\text{Ra}^{221}$  there is no obvious rotational pattern present although the levels at 324 and 362 kev appear to form the base members

to a rotational band. As previously stated (Section 2-e) there are no levels present which can be interpreted as higher excited rotational members. In  $\text{Rn}^{217}$  there is certainly no rotational pattern present. The spectrum becomes much simpler in  $\text{Po}^{213}$ , and in  $\text{Pb}^{209}$  the typical single particle spectrum is exhibited.

Attempts have been made to interpret energy levels of nuclides on the border of the transition region in terms of the Bohr-Mottelson theory. Levels in  $\text{Ac}^{225}$ ,  $\text{Ac}^{223}$ , and  $\text{Ra}^{223}$  have apparently been partially explained.<sup>8,29,36</sup> In these cases the rotational constants range in value from 6 to 9 kev. Some of the levels in  $\text{Fr}^{221}$  and  $\text{Ra}^{223}$  have also been interpreted in this way but here the rotational constants are 13.3 and 17.4 kev, respectively.<sup>37</sup> The latter value seems very high in view of the fact that in  $\text{Ra}^{222}$  and  $\text{Ra}^{224}$  it is 20 and 14.5 kev, respectively, and rotational constants are known to lie lower for odd mass nuclides, at least in all other cases studied.

The levels in  $\text{Ra}^{225}$  are somewhat similar to those in  $\text{Ra}^{221}$  in that 58 percent of the alpha population goes to a state at 214.5 kev with a hindrance factor of 1.5. Since the ground state of  $\text{Th}^{229}$  has  $K = I = 5/2$ , then favored alpha decay implied by the small hindrance factor strongly suggests the same assignment for the 214.5 state. However, there are no obvious rotational levels based on this state. It was suggested<sup>37</sup> that a strong interaction between the levels is responsible for the apparent absence of rotational structure.\* If this is true, then this could possibly explain the lack of rotational level structure in  $\text{Ra}^{221}$ . In this regard it should be noted that considering the Nilsson diagram, the only levels in the vicinity which would interact would be the  $5/2+$  (633) and  $3/2+$  (631). The levels are about 300 kev in  $\text{U}^{233}$  and possibly only 71 kev in  $\text{Th}^{229}$  (Section C). It is possible then, that the levels are close enough in  $\text{Ra}^{225}$  and  $\text{Ra}^{221}$  to interact appreciably and thus distort the rotational level spacings. This may well be the case since  $\text{Ra}^{223}$  appears to exhibit a rotational level pattern. It is obvious that more experimental information is needed to decide whether or not the strong coupling model can explain the results in odd-mass radium isotopes.

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\* The effect referred to here was first worked out by Kerman.

The complexity of the level structure in  $\text{Rn}^{217}$  or  $\text{Rn}^{219}$  and the absence of a rotational pattern clearly indicate the "softness" of the nucleus to distortion on the equilibrium spheroidal shape which illustrates the need for theoretical development in this region of transition from spheroidal to spherical.

## B. Uranium-228 Series

The decay of a 9.3 minute uranium isotope produces a five-membered series which is collateral with the thorium (4n) family (Figure 30). Since this series joins the main family at  $\text{Po}^{212}$  and also decays to  $\text{Pa}^{228}$ , this uranium isotope has been assigned the mass number 228.<sup>23</sup>

The alpha particle energies (Table X) were determined with an ionization chamber by Meinke<sup>23</sup> who produced the  $\text{U}^{228}$  by an  $(\alpha, 8n)$  reaction on  $\text{Th}^{232}$  and by Orth<sup>39</sup> who obtained the activity from the alpha decay of  $\text{Pu}^{232}$  made by an  $(\alpha, 7n)$  reaction on  $\text{U}^{235}$ . Figure 30 and Table X summarize all the data which have been previously reported.<sup>23,24,39</sup>

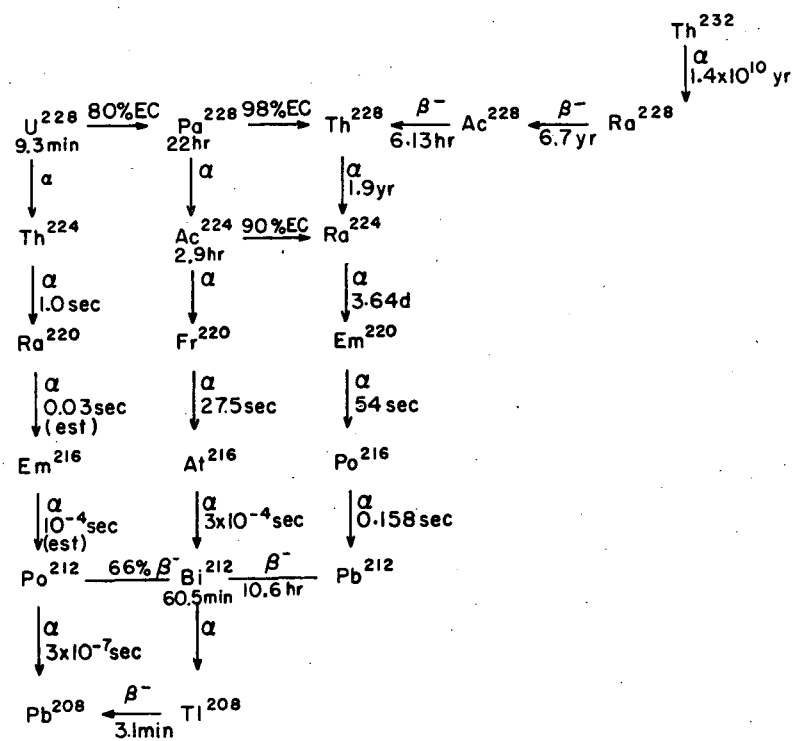
TABLE X

| Activity          | Alpha Particle Energies (Mev) |                 |                 |
|-------------------|-------------------------------|-----------------|-----------------|
|                   | Meinke                        | Orth            | This work       |
| $\text{U}^{228}$  | 6.72                          | $6.67 \pm 0.02$ | $6.68 \pm 0.01$ |
| $\text{Th}^{224}$ | 7.20                          | $7.13 \pm 0.02$ | $7.17 \pm 0.01$ |
| $\text{Ra}^{220}$ | 7.49                          | $7.43 \pm 0.03$ | $7.45 \pm 0.01$ |
| $\text{Rn}^{216}$ | 8.07                          | $8.01 \pm 0.03$ | $8.04 \pm 0.01$ |

Because of the short half-lives of the members of this series, separation of the daughter activities was not feasible. Therefore, the data had to be obtained on the series as a whole, and electronic methods had to be relied upon to make the "separations". This then influences the mode of presentation — the alpha particle and gamma ray data will be presented for the whole family (Sections 1 and 2) but the analysis and interpretation will be made with respect to each member separately (Section 4).

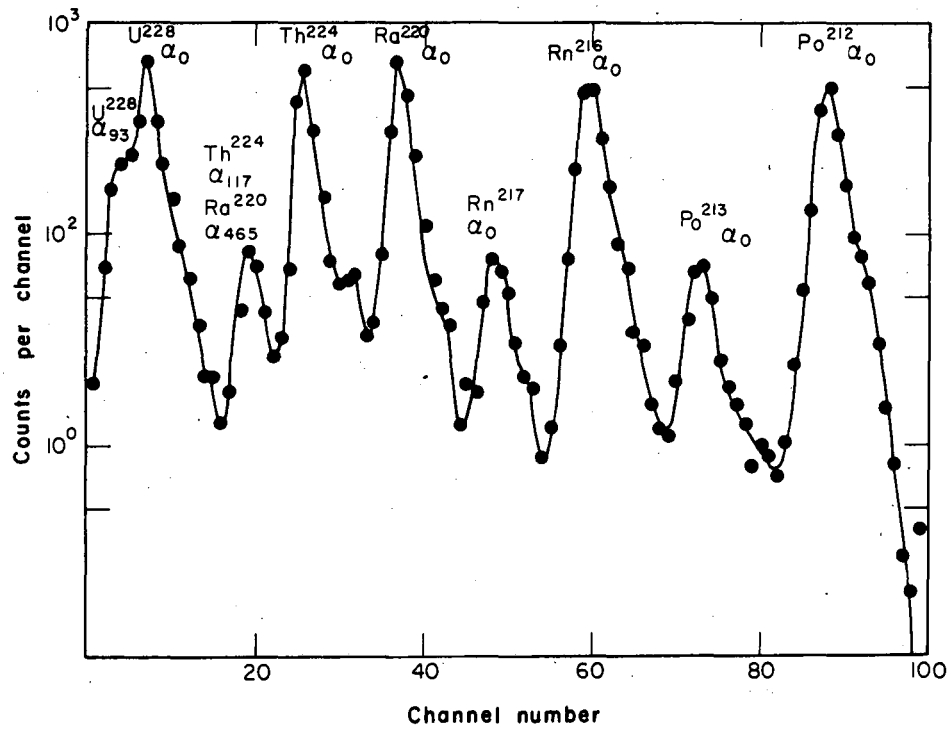
### 1. Alpha spectra

Figure 31 is a typical alpha spectrum of the whole  $\text{U}^{228}$  family taken with the alpha grid chamber. This spectrum was obtained from an 8-minute count with the uranium sample collimated with the 100-count electroformed screen previously mentioned (Section II, A-2); the count was started 18 minutes after the end of a 20 minute bombardment. Peaks due to the five members of the decay series are clearly



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Fig. 30. Thorium family and collateral  $U^{228}$  family.



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Fig. 31. Alpha particle spectrum of the  $U^{228}$  family using a 100-count mesh collimator.

in evidence. Their energies are tabulated in Table X. ( $\text{Po}^{212}$  is not included since its alpha particle energy is well known).<sup>32</sup> The energies are seen to be in general agreement with the previous measurements. The decay of the peaks was followed by taking a series of 8-minute counts and was found to decay with the 9-minute half-life of the  $\text{U}^{228}$  parent.

Beside the  $\text{U}^{228}$  family, the presence of the  $\text{U}^{229}$  family is obvious. The  $\text{Rn}^{217}$  and  $\text{Po}^{213}$  groups are clearly visible; the higher energy groups of  $\text{Ra}^{221}$  and  $\text{Th}^{225}$  although less obvious are also present, occurring in the same energy region as  $\text{U}^{228}$ .

From this spectrum two new alpha groups are seen; one occurring as a shoulder on the low energy side of the  $\text{U}^{228}$  peak and one occurring at 0.18 Mev lower in energy than the  $\text{Th}^{224}$  peak. These also were observed to decay with the 9-minute half-life. The hump occurring in between the  $\text{Th}^{224}$  and  $\text{Ra}^{220}$  peaks was found to be spurious since it did not appear in other spectra (Figure 32).

In order to investigate the possibility of fine structure, the energy region between the  $\text{U}^{228}$  and  $\text{Ra}^{220}$  alpha groups was examined in greater detail (Figure 32). Beside the ground state groups three other peaks are apparent at 6.60, 6.77, and 6.99 Mev. Two of the groups decayed with the 9-minute half-life and were attributed to the decay of the  $\text{U}^{228}$  series; the one at 6.77 Mev decayed with a 1-hour half-life and was due to the  $\text{Ra}^{221}$  and  $\text{Th}^{225}_{\alpha_0}$  groups.

## 2. Coincidence Spectra

The presence of the  $\text{U}^{229}$  family with its intense gamma rays rendered total alpha-gamma coincidence techniques useless for the detection of the presumably relatively weak gamma rays of the  $\text{U}^{228}$  family. Therefore, the alpha gate was restricted to those energy regions where no intense excited state alpha groups from the  $\text{U}^{229}$  family appeared.

In all the coincidence spectra discussed the ring collimator was used. Since the samples used were too active for best resolution in the grid chamber, the peaks were broadened in the gamma-alpha coincidence spectra.

### a. Thorium-224 and Radium-220

#### (1) 6.75 - 7.5 Mev Alpha-Gamma Coincidence Spectra:

The alpha energy band from 6.75 to 7.5 Mev was used as a gate and the coincident gamma spectrum obtained is shown in Figure 33.

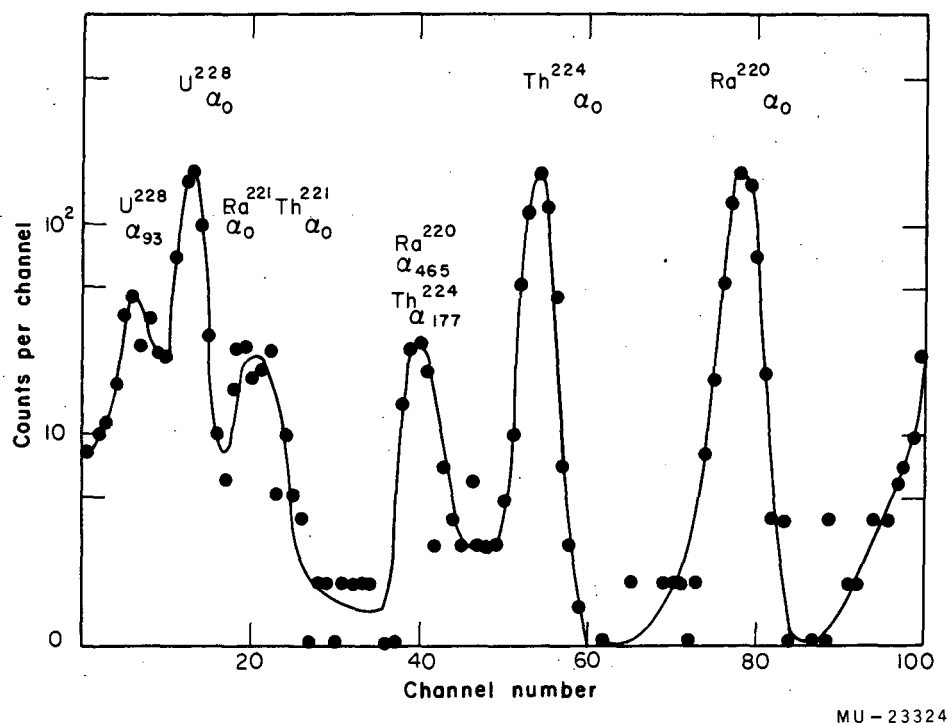
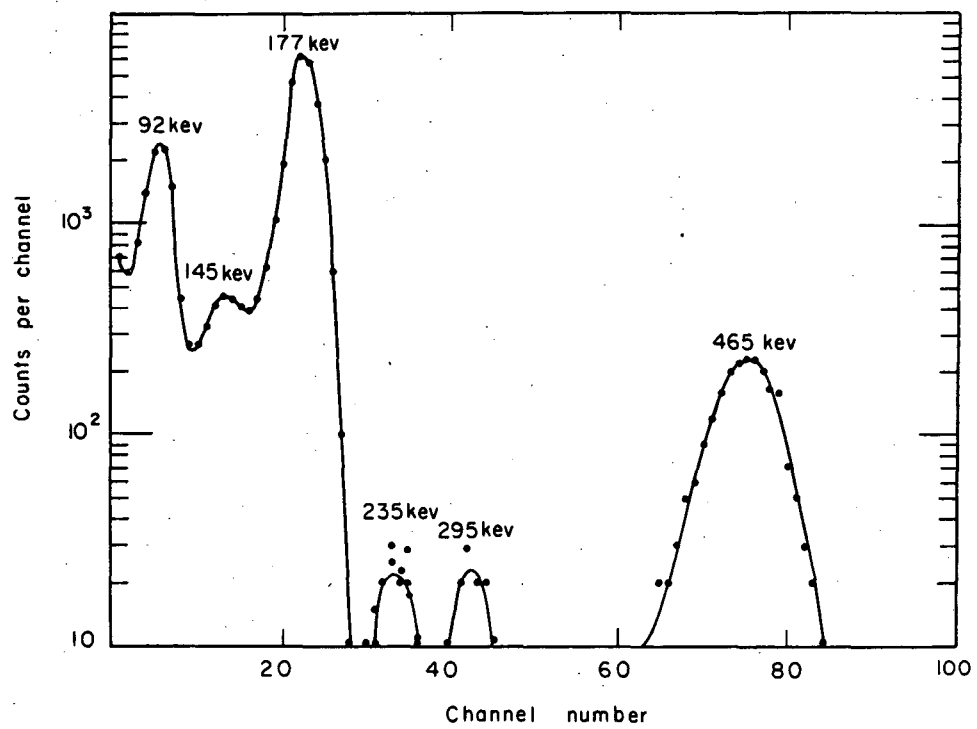


Fig. 32. Alpha particle spectrum of  $U^{228}$ ,  $Th^{224}$ , and  $Ra^{220}$  using a ring collimator.



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Fig. 33. 6.75 - 7.5 Mev Alpha particle - gamma ray coincidence spectrum of  $\text{Th}^{224}$  and  $\text{Ra}^{220}$ .

Gamma rays with the following intensities were seen: 92 kev (3%), 140 kev (0.5%), 177 kev (9%), 235 kev (0.1%), 295 kev (0.1%), and 465 kev (1%).

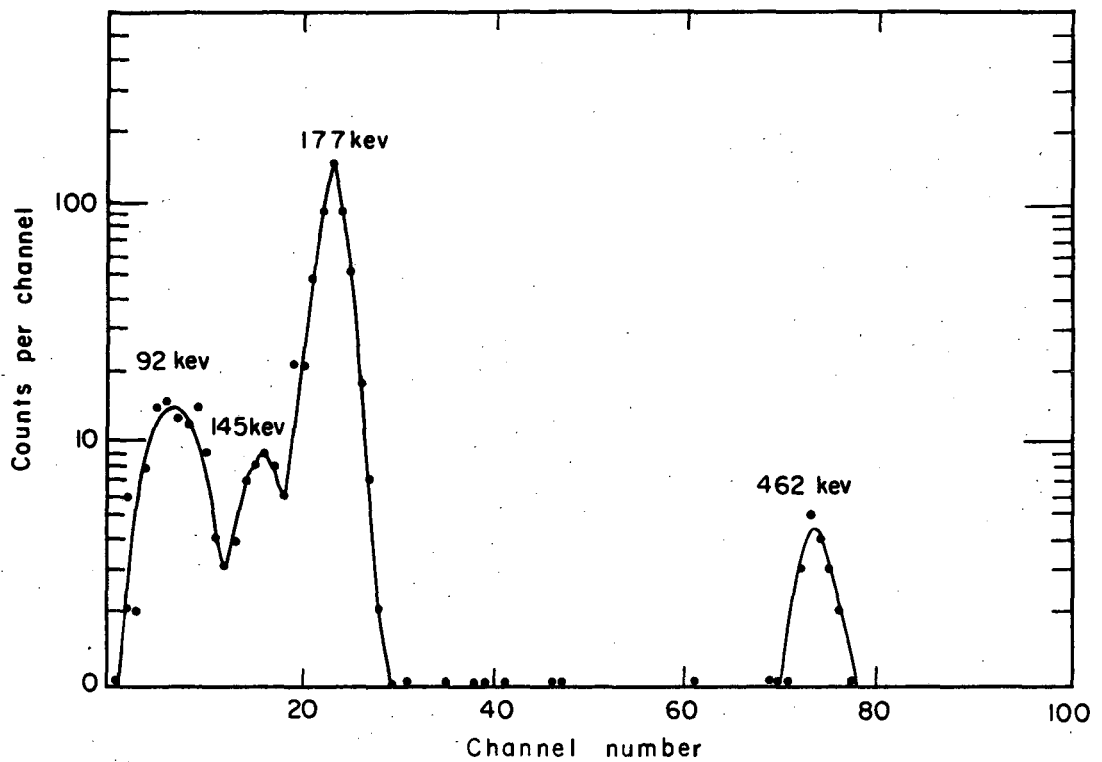
(2) 6.99 Mev Alpha-Gamma Coincidence Spectrum:

The energy of the 177 kev gamma ray is in agreement with the energy difference between the 7.17 and 6.99 Mev alpha groups, which suggested that the 6.99 Mev group was associated with the decay of  $\text{Th}^{224}$ . In order to show this, the 6.99 Mev alpha group was gated on; Figure 34 shows the gamma spectrum obtained in coincidence. The following gamma ray energies and intensities were observed: 90 kev (2%), 145 kev (0.7%), 177 kev (9%), and 462 kev (1%). This proved that the 177 kev transition was associated with the de-excitation of the level populated by the 6.99 Mev group to the ground state of  $\text{Th}^{224}$ . From the ratio of intensities of K x-rays to the 177 kev gamma ray, a K-conversion coefficient of 0.22 was indicated. This is in good agreement with an E2 K-conversion coefficient of 0.20. From the intensity of the two radiations and using the L and M conversion coefficients, the population to the 177 kev level was calculated to be 18 percent in good agreement with the value of 19 percent determined from the alpha spectra (Table XI).

The 462 kev gamma ray must come from de-excitation of a level also populated by an alpha group of about 6.99 Mev. Since this cannot come from  $\text{Th}^{224}$  it must be due to  $\text{Ra}^{220}$ . The energy agrees well with the energy difference of the alpha particles to the two states (6.99 and 7.45).

(3) Gamma-Alpha Coincidence Spectra:

The presence of three gamma rays has yet to be explained. Gating on all gamma rays, the spectrum of alpha particles counted in coincidence was obtained. There was an intense group at 6.99 Mev and a complex group at lower energies. The  $\alpha_0$  groups of  $\text{Th}^{225}$  and  $\text{Ra}^{221}$  were not in coincidence and therefore did not appear in the spectrum. At their same energy however (about 6.73 Mev), there was a distinct high energy shoulder on the  $\text{Ra}^{221} - \text{Th}^{225}$  excited state complex groups. In order to better resolve this shoulder from the complex  $\text{U}^{229}$  family groups, the gate was set to accept only those gamma rays having energies in excess of 180 kev; the resulting coincidence spectrum is shown in Figure 35. The group at 6.73 Mev is now well separated from the  $\text{U}^{229}$



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Fig. 34.  $\text{Th}^{224}\alpha_{177}$  and  $\text{Ra}^{220}\alpha_{465}$  - gamma ray coincidence spectrum.

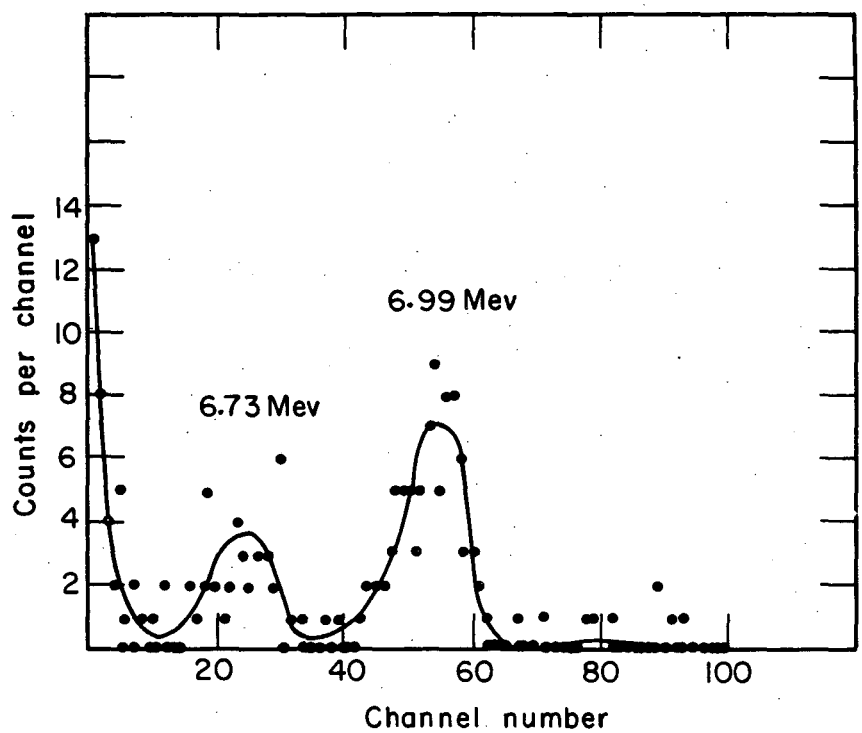
Table XI. Alpha Groups of the  $U^{228}$  Family

| Nuclide    | Alpha Particle<br>Energy (Mev) | Abundance<br>%  | Excited State<br>Energy (kev) | Hindrance<br>Factor <sup>c</sup> |
|------------|--------------------------------|-----------------|-------------------------------|----------------------------------|
| $U^{228}$  | $6.68 \pm 0.01$                | 70              | 0                             | 1.0                              |
|            | 6.59                           | 29 $\pm 4$      | $93 \pm 4$                    | 1.0                              |
|            | $6.44^a$                       | $0.7 \pm 0.3^b$ | $246 \pm 3$                   | 11                               |
|            | $6.40^a$                       | $0.5 \pm 0.2^b$ | $280 \pm 6$                   | 10                               |
| $Th^{224}$ | $7.17 \pm 0.01$                | 79              | 0                             | 1.0                              |
|            | 6.90                           | 19 $\pm 2$      | $177 \pm 3$                   | 1.0                              |
|            | 6.77                           | $1.5 \pm 0.6^b$ | $410 \pm 4$                   | 2                                |
|            | 6.70                           | $0.5 \pm 0.3^b$ | $475 \pm 5$                   | 3                                |
| $Ra^{220}$ | $7.45 \pm 0.01$                | 99              | 0                             | 1.0                              |
|            | $6.90^a$                       | 1 $\pm 0.4^b$   | $465 \pm 4$                   | 2                                |
| $Em^{216}$ | $8.04 \pm 0.01$                | 100             | 0                             | 1.0                              |

<sup>a</sup> Existence of groups and their energy inferred from gamma ray data.

<sup>b</sup> Abundance obtained from gamma ray intensities.

<sup>c</sup> Calculations based on half-lives of 9.1 min, and 1.0, 0.023, and  $4.5 \times 10^{-5}$  sec starting with  $U^{228}$  for which an alpha branching ratio of 0.95 was used.



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Fig. 35. Gamma ray  $> 180$  keV - alpha particle coincidence spectrum of  $\text{Th}^{224}$  and  $\text{Ra}^{220}$ .

family groups. The group at 6.99 Mev is also apparent but greatly reduced in intensity because it is in coincidence with only the 465 kev gamma ray.

(4) 6.73 Mev Alpha-Gamma Coincidence Spectrum:

The coincidence spectrum obtained by gating on the 6.73 Mev alpha group is shown in Figure 36 which is a compilation of the results of the four best experiments. The energies and approximate intensities are: 88 kev (0.9%), 145 (0.2%), 177 (0.6%), 235 kev (0.4%), 295 kev (0.3%), and 410 kev (0.8%). Since the gating alpha group is higher in energy than the  $U^{228}\alpha_0$ , these radiations cannot be associated with its decay. They cannot be associated with the  $Ra^{220}$  decay either because the alpha intensities to the states are much too high; also at least some decay would have to occur to the level at 465 kev on the basis of gamma ray energy consideration in which case the 465 kev radiation would have been observed. Therefore, it seems certain that these radiations are associated with  $Th^{224}$  decay.

(5) 6.65 - 7.5 Mev Alpha-Gamma Coincidence Spectra:

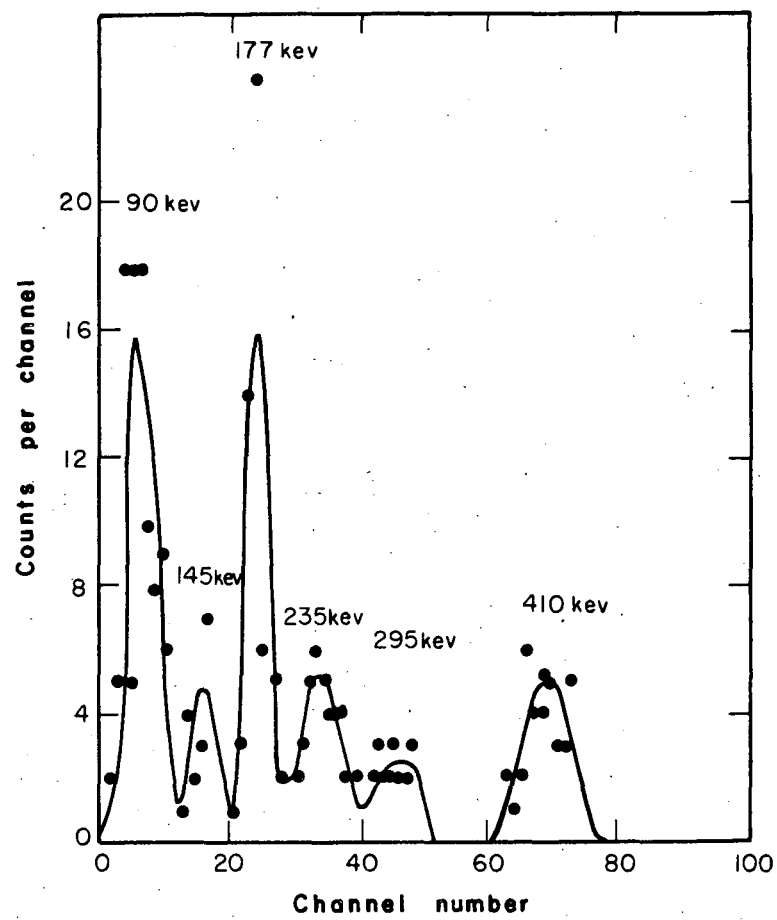
From the intensities of the 235, 295, and 410 kev gamma rays obtained in the 6.75 - 7.5 Mev and 6.73 Mev alpha-gamma coincidence experiments (Section 1 and 4) are seen to be different. In fact, the 410 kev gamma ray was not seen in the 6.75 - 7.5 Mev coincidence experiment. The reason is that the 6.73 Mev alpha group was partially discriminated out of the gating region and therefore the gamma ray intensities associated with the de-excitation of the level populated by the group are lower. The experiment was repeated with the threshold lowered slightly to accept all of the pulses of the 6.73 Mev alpha group. The gamma ray energies and intensities obtained were: 177 kev (9%), 235 kev (0.4%), 295 kev (0.4%), 410 kev (0.8%), and 465 kev (1%) in good agreement with the previous values (Sections 1, 2, and 4).

b. Uranium-228

From the alpha spectra (Figures 31 and 32, and Section a) a group having an energy 6.59 Mev was observed. From its intensity and energy it must be associated with the decay of  $U^{228}$ , populating a state about 93 kev above the ground state.

(1) 6.39 - 6.45 Mev Alpha-Gamma Coincidence Spectra:

By analogy with other even-even nuclei in this region it was expected that the decay of  $U^{228}$  should populate two higher excited states in  $Th^{224}$ ,



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Fig. 36. 6.73 Mev alpha particle - gamma ray coincidence spectrum of  $\text{Th}^{224}$ .

one at an energy approximately 3.0 times the first excited state energy (Section 3, Figure 42) giving an energy level at 280 keV above the ground state. The energy of the alpha group to be used as a gate to see the gamma rays would thus be about 6.40 MeV. Now  $\text{Th}^{225}$  has intense alpha groups at 6.47 and 6.44 MeV which could cause serious interference, especially if the sought for group was at an energy higher than 6.40 MeV. Four experiments were performed in which the region from 6.45 to 6.35 MeV was traversed in about 40 keV intervals. In the gate region from 6.39 to 6.45 MeV three gamma rays were observed in the coincidence spectrum which were above the intensity of the gamma rays of  $\text{Th}^{225}$ . The spectrum, with the  $\text{Th}^{225}$  contribution subtracted is shown in Figure 37. The gamma ray energies and intensities were: 90 keV (0.4%), 152 keV (0.2%), 187 keV (0.3%), 246 keV (0.4%).

#### (2) L X-ray-Alpha Coincidence Experiment:

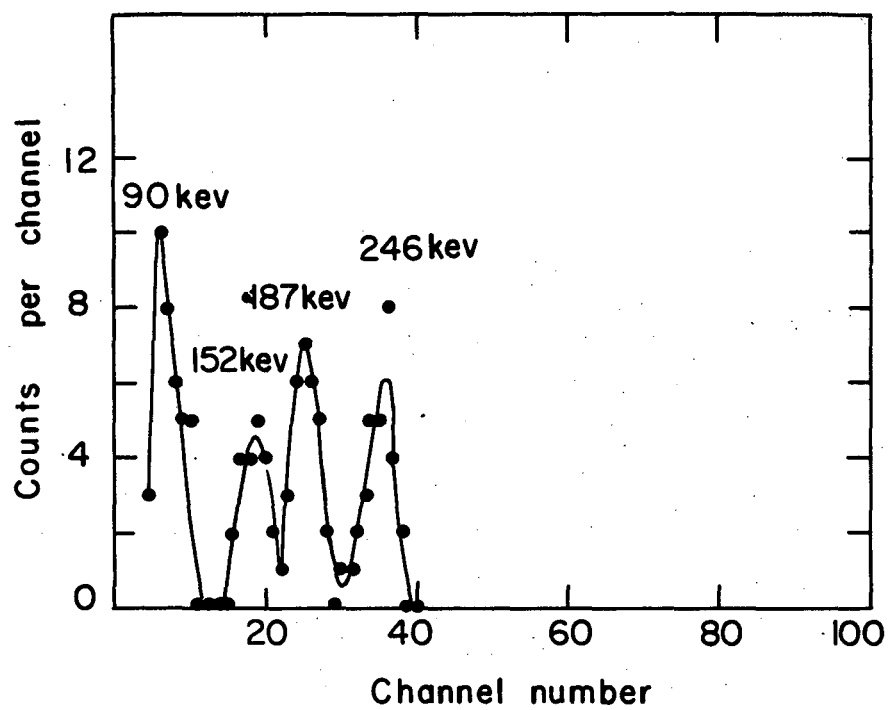
In order to try to observe the alpha groups populating these higher excited states an L x-ray-alpha coincidence experiment was performed. It was argued that since the levels must partially de-excite through the 93 keV level which decays in part by L electron conversion, then these groups might be observed. (Gamma-alpha coincidence experiments were not possible because of the high intensity gamma rays de-exciting levels populated by about the same energy alpha groups in  $\text{Th}^{225}$  decay.) Several L x-ray-alpha experiments were run but because of the small number of events no alpha groups were seen above those of the  $\text{U}^{229}$  family. Therefore the existence of these excited states is not conclusively proven but strongly suggested by the gamma rays.

The alpha particle and gamma ray data obtained in the previous sections are summarized in Tables XI and XII. An interpretation of the results will be given in Section 4.

### 3. Half-Life and Branching Ratio Determinations

#### a. Uranium-228

The half-life of  $\text{U}^{228}$  has been reported as  $9.3 \pm 0.5$  minutes and the alpha branching ratio as about 80 percent.<sup>23</sup> Using these values and an electron capture decay energy of 0.34 MeV (obtained from closed cycle decay using the alpha energies of Table XI) a log ft value of 4.0 is obtained. This value seems extremely low, because even in the case of allowed unhindered decay the log ft is at least 4.5. Also, this type of



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Fig. 37. 6.39 - 6.45 Mev alpha particle - gamma ray coincidence spectrum of  $U^{228}$ .

TABLE XII

| Gamma Rays of $U^{228}$ Family |              |                         |
|--------------------------------|--------------|-------------------------|
| Nuclide                        | Energy (kev) | Abundance Photons/Alpha |
| $U^{228}$                      | $152 \pm 3$  | $0.002 \pm 0.0005$      |
|                                | $187 \pm 3$  | $0.003 \pm 0.001$       |
|                                | $246 \pm 3$  | $0.004 \pm 0.001$       |
| $Th^{224}$                     | $90 \pm 2$   | $0.02 \pm 0.01$         |
|                                | $177 \pm 2$  | $0.09 \pm 0.02$         |
|                                | $235 \pm 3$  | $0.004 \pm 0.002$       |
|                                | $297 \pm 3$  | $0.003 \pm 0.001$       |
|                                | $410 \pm 3$  | $0.008 \pm 0.003$       |
| $Ra^{220}$                     | $465 \pm 4$  | $0.01$                  |

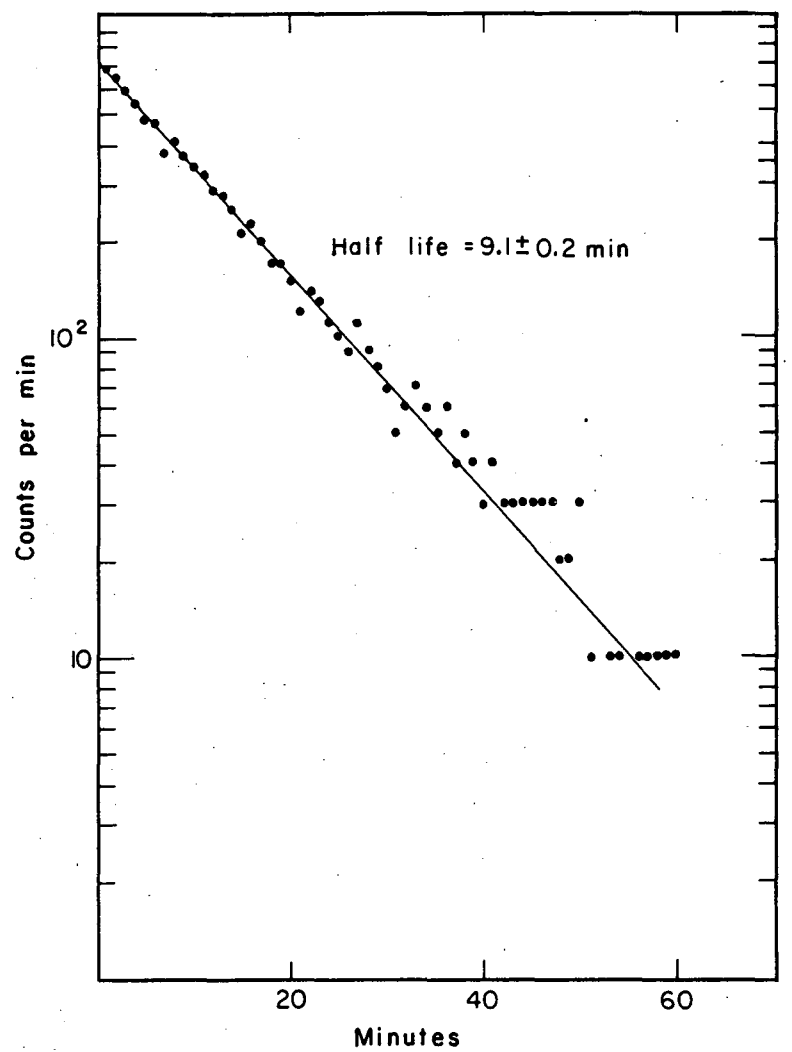
decay is unlikely in view of the fact that an even-even nucleus is decaying to an odd-odd. Therefore, it appeared that either the half-life or the branching ratio (or both) were in error.

(1) Half-Life Determination:

The half-life was measured by gating in the alpha pulses from the decay of  $Po^{212}$  (which was in equilibrium with  $U^{228}$ ), with a single-channel pulse height analyzer. After inversion the output pulse was fed into a scalar to which a traffic counter was attached. The rate of decay of the  $Po^{212}$  thus gave a direct measure of the  $U^{228}$  half-life. In this way the contribution to the decay curve from the presence of  $U^{229}$  and  $U^{230}$  was reduced to an insignificant amount in which case the decay curve had essentially no "tail". A typical curve is shown in Figure 38. Two measurements gave a half-life of  $9.1 \pm 0.2$  minutes, in good agreement with the previously reported value.

(2) Branching Ratio Determination:

The branching ratio was determined as follows: the same chemical procedures as outlined in Section II, C-2, were used to extract the evaporation of the ether phase, the residual solution was transferred to a 1 ml volumetric flask containing  $Pa^{231}$  tracer and then diluted up to the



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Fig. 38. Decay curve of  $U^{228}$ .

mark by the addition of 8 M HCl. From this was taken a 0.025 ml aliquot and the remainder passed through a small anion exchange column. Then 1 ml of 8 M HCl - 0.5 M HF was passed through the column stripping off the Pa. The completion was then counted on the alpha grid chamber from time,  $t$  to  $t'$ . Knowing the half-life and the number of  $U^{228}$  counts obtained from  $t$  to  $t'$ , the amount of  $U^{228}$  on the column at time  $t_0$  could be calculated.

Three  $U^{228}$  half-lives after  $t_0$ , another milliliter of 8 M HCl - 0.5 M HF was passed through the column to strip off the Pa again. The plates containing the activity from the first and second Pa elutions were counted for both  $Pa^{231}$  and  $Pa^{228}$ . A comparison of the  $Pa^{231}$  counts in each sample gave the efficiency of the elution. From this number, and the counts of  $Pa^{228}$  on the first plate, the number of  $Pa^{228}$  counts on the second plate not due to the decay of  $U^{228}$  from  $t_0$  could be determined. From the data obtained an upper limit of 5 percent could be set for the electron capture branching ratio of  $U^{228}$ .

Using this number, then  $\log ft \geq 4.7$ , which still seems low but is more reasonable. This also gives a half-life-energy relation in better than the 80 percent alpha branching ratio.<sup>41</sup>

#### b. Radium-220 Half-Life Measurement

The half-life was measured using the apparatus and technique described in Section II, C-2. The curve obtained from a typical experiment is shown in Figure 39. The half-life obtained from several experiments gave the value  $0.023 \pm 0.05$  seconds. From the half-life-energy relation a somewhat shorter half-life was calculated (but within the lower limit of error of the experimental value).

#### c. Radon-216 Half-Life Determination

The half-life of  $Rn^{216}$  was measured using the time-to-pulse-height converter system described in Section II, C-1. The  $Ra^{220}$  alpha pulses triggered the system and the  $Rn^{216}$  alpha pulses were used as signal pulses. A typical decay curve obtained with a sweep time of 1 millisecond is shown in Figure 40. Five determinations using sweep times of 0.5 and 1 millisecond gave the value of  $4.5 \pm 0.5 \times 10^{-5}$  seconds. The half-life-energy relation is in good agreement with that established by other even-even radon isotopes.<sup>41</sup>

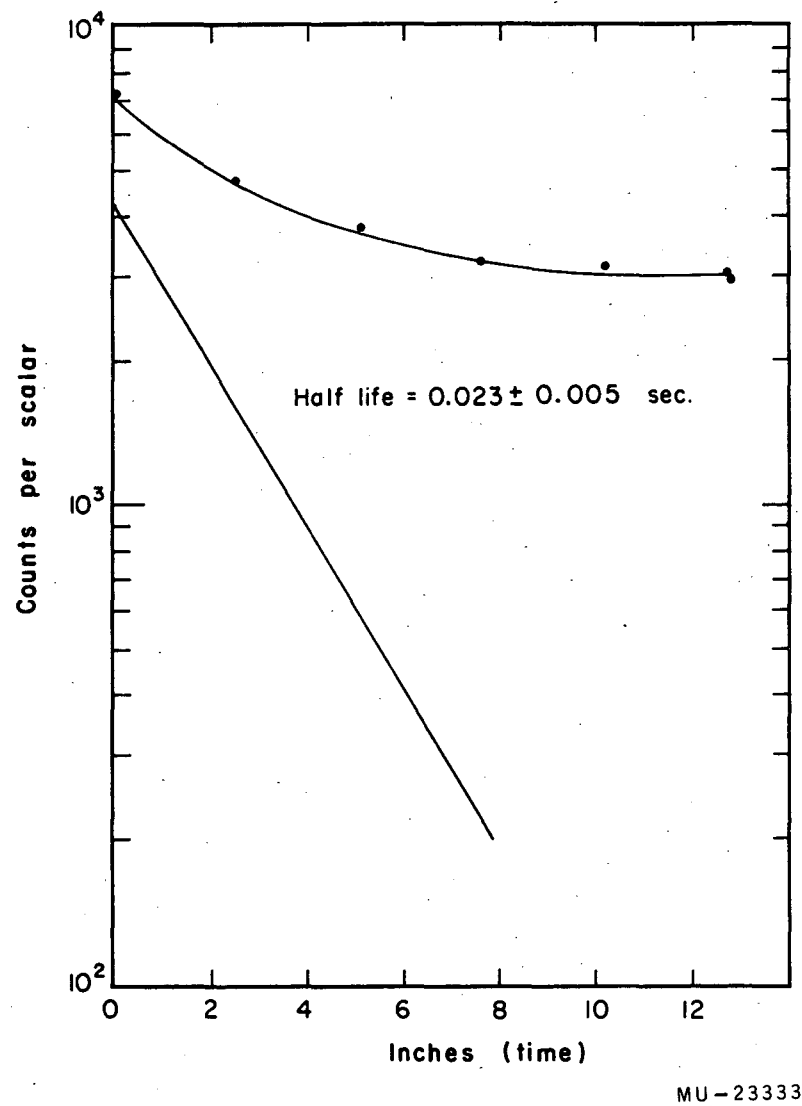


Fig. 39. Decay curve of  $\text{Ra}^{220}$ .

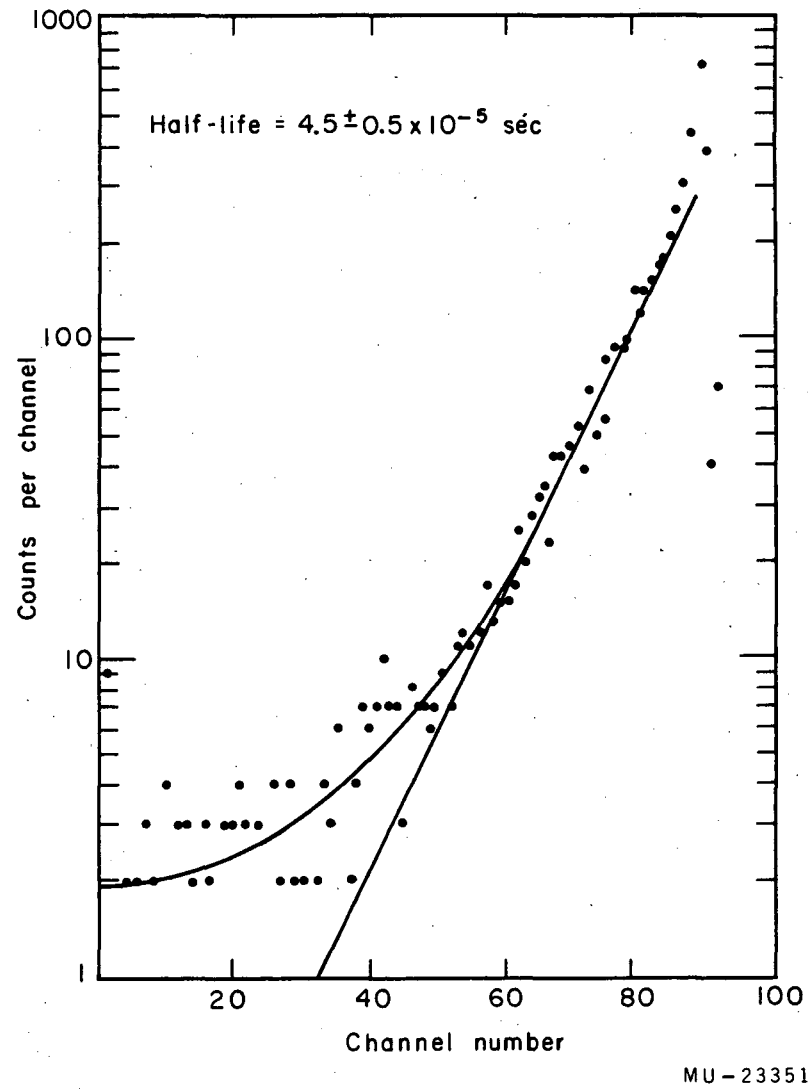


Fig. 40. Delay coincidence curve of  $\text{Rn}^{216}$ .

#### 4. Discussion and Interpretation

The energy levels observed in this study cannot be assigned definite spins and parities on the basis of the experimental results obtained in most cases. The assignments will be made utilizing the data available on other even-even nuclides in this region, and theory to some extent. Before beginning the analysis, the theory will be reviewed very briefly.

##### a. Nuclear Models

The axially symmetric nuclear model of Bohr and Mottelson has been very successful in explaining spectra in regions far removed from closed shells where nuclei have large permanent deformations from the spherical shape. For even-even nuclei the low lying levels expected are  $0+$ ,  $2+$ ,  $4+$ , ... having the well known energy spacing given by the formula

$$E = A I (I + 1) - B I^2 (I + 1)^2 \quad (5)$$

where  $A = \hbar^2/2\mathcal{I}$  ( $\mathcal{I}$  is the moment of inertia). The second term corrects for the deviation of the theoretical from the experimental value due to rotation-vibration interaction. In regions of large deformation  $B \approx 0$ . As the regions near closed shells are approached the value of  $B$  rapidly increases. Finally, the place is reached where the spectra can no longer be explained for any value of  $A$  and  $B$  in Equation (5). This transition region from rotational to vibrational states has been estimated to occur when the energy of the first-excited  $2+$  state is

$$E(2+) \approx 13 \hbar^2/\mathcal{I} \text{ rig} \quad (6)$$

where  $\mathcal{I} \text{ rig}$  is the moment of inertia associated with the rigid rotation of a sphere. Upon evaluation, the transition is found to occur for mass numbers of about 188 and 222. In a comparison of the experimental data with theory, it was found that for no choice of  $A$  and  $B$  could Equation (5) be made to agree with the data for  $\text{Os}^{188,190,192}$  and  $\text{Pt}^{192}$ .<sup>43</sup> Mallman<sup>44</sup> has made a study of the agreement between experiment and theory and has found that the rotational region is confined to values of  $R_4 \geq 3.27$  ( $R_4$  is the ratio of the energies of the  $4+$  to the  $2+$  states) since deviations begin to occur at this point. Therefore the Bohr-Mottelson model cannot be applied with success to the near harmonic nuclei and in

particular to this study since  $R_4$  for mass number 224 ( $\text{Ra}^{224}$ ) is already 3.02 (Section 2, Figure 45).

Using the assumption that nuclei possess equilibrium shapes which are not necessarily axially symmetric, Davydov, Filippov and Rostovsky<sup>4,5,45</sup> have developed a theory to treat the rotational levels of even-even nuclei. The energy states are considered to correspond to rotation of the nucleus as a whole with no change in its internal shape. Energy levels are calculated in terms of  $\gamma$  which is a measure of the deviation of the shape from axial symmetry;  $\gamma$  varies between 0 and  $\pi/3$ . For  $\gamma = 0$  the nucleus is a prolate spheroid and for  $\gamma = \pi/3$  it is an oblate spheroid. For  $\gamma \neq 0$  or  $\pi/3$  the nucleus is regarded as an asymmetric top. Since the rotational states and the probabilities of electromagnetic transitions are the same for  $\gamma$  as  $\pi/3 - \gamma$ ,  $\gamma$  ranges between 0 and  $\pi/6$ .

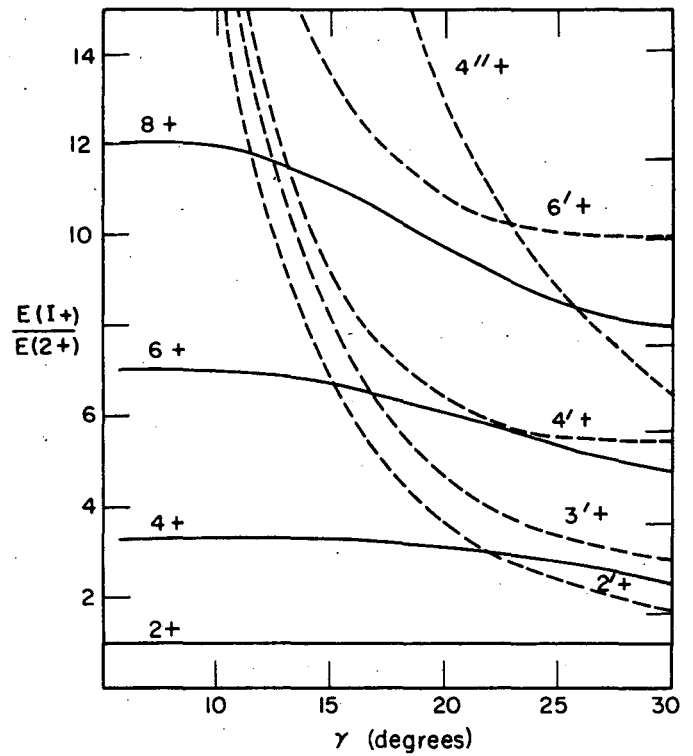
Besides the levels of spin and parity  $0+$ ,  $2+$ ,  $4+$ , ... given by the Bohr-Mottelson theory, the theory of non-axial nuclei yields other rotational levels with even and odd spins. The energies of these states can no longer be obtained by a simple  $I(I+1)$  formula but are now functions of  $\gamma$ . The quantity  $\gamma$  is usually computed from the ratio of the energy of the second excited  $2+$  state ( $E_2'$ ) to that of the first  $2+$  state ( $E_2$ )

$$R_2 = \frac{E_2'}{E_2} = \frac{3 + [9 - 8 \sin^2(3\gamma)]^{1/2}}{3 - [9 - 8 \sin^2(3\gamma)]^{1/2}}. \quad (7)$$

Knowing  $\gamma$ , the energies of the other states can be determined. (Of course  $\gamma$  can also be computed from the energies of any two "old" rotational levels (i.e.,  $2+$ ,  $4+$ , ...) but then corrections must be made for vibration-rotation interaction.)

Figure 41 shows the dependence of the level energies on  $\gamma$ . It is seen that in the vicinity of  $\gamma = 0$  the "new" rotational levels are very high in energy and the spectrum is the same as the axially-symmetric one. However, as  $\gamma$  increases these new levels decrease in energy such that at  $21.5^\circ$  the  $2'+$  level crosses below the  $4+$  level.

There have been several papers published comparing the asymmetric rotor theory with experimental data.<sup>43,46-48</sup> Very briefly the results in the region concerned within this research are ( $R_4$  from  $8/3$  to  $3.27$ ):



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Fig. 41. Diagram showing the variation of the ratios of energy levels with  $\gamma$ .

all ratios are in fairly good agreement with theory except for  $\text{Sm}^{152}$ ,  $\text{Gd}^{154}$ , and  $\text{Os}^{186}$ . (For  $\text{Os}^{186}$  there is some question about the spin assignments), for  $3.27 < R_4 < 3.33$  the non-axial and axial theories are equivalent. The theory is also in qualitative agreement with the observed trends of the ratio of reduced E2 transition probability ratios from the second excited  $2^+$  state.

Recently the theory has been modified to take into account vibration-rotation interaction. In this treatment only the  $\beta$  vibrations were taken into account and it was assumed that the nucleus was rigid with respect to a change in the equilibrium value of the parameter  $\gamma$ . The energy levels are given in terms of two parameters,  $\gamma$ , the "non-axiality" parameter, and  $\mu$ , the "non-adiabaticity" parameter which are determined from the energies of three levels:  $E_2$ ,  $E_2'$ , and one other.

The value  $R_2$  is 2 for  $\gamma = 30^\circ$  and  $\mu = 0$  (the adiabatic condition). If the coupling between vibration and rotation is taken into account, this ratio decreases to 1.6 for  $\mu = 1$ . Likewise for  $\gamma = 0^\circ$  the ratio  $R_4$  can vary from  $10/3$  ( $\mu = 0$ ) to  $2.25$  ( $\mu = 1$ ); for  $\gamma = 30^\circ$  the ratio becomes  $8/3$  ( $\mu = 0$ ) and  $1.95$  ( $\mu = 1$ ). Using these two parameters good agreement was obtained with  $\text{Sm}^{152}$  ( $\mu = 0.37$ ) and  $\text{Gd}^{154}$  ( $\mu = 0.36$ ) which is noted above gave poor agreement with the one parameter theory. This non-axially symmetric theory then appears to be applicable to the region under investigation, or at least has a useful correlative function.

#### b. Interpretation

A considerable amount of data are now available on the states in even-even nuclei in the region above lead. Figure 42 shows the ratios of the energies of  $4^+$  and  $2^+$  to  $2^+$  states on both sides of the closed shell at  $\text{Pb}^{208}$  as a function of mass number.\* Figure 43 gives a similar plot for the  $1^-$  states and also the energies of first excited  $2^+$  states in the trans-lead region. These figures will be referred to frequently throughout the following sections of the paper in analyzing the data.

##### (1) Uranium-228:

The data (Sections 1 and 2b) indicate the existence of an excited state at 93 kev. This group could not be used as a gate because of the

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\* Data taken from references 24, 43, 50, and 51.

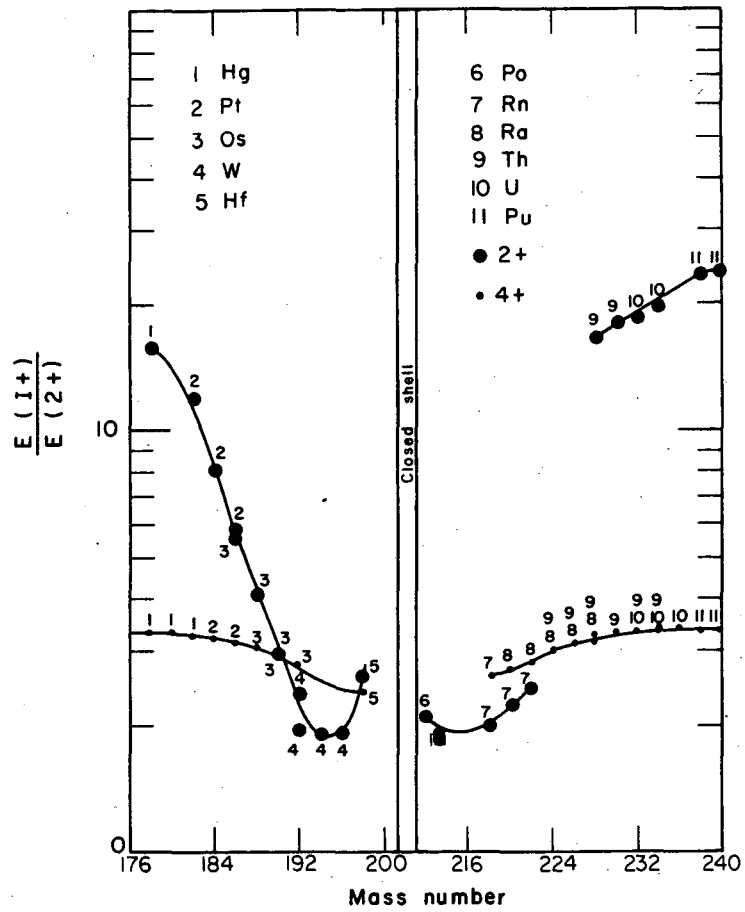
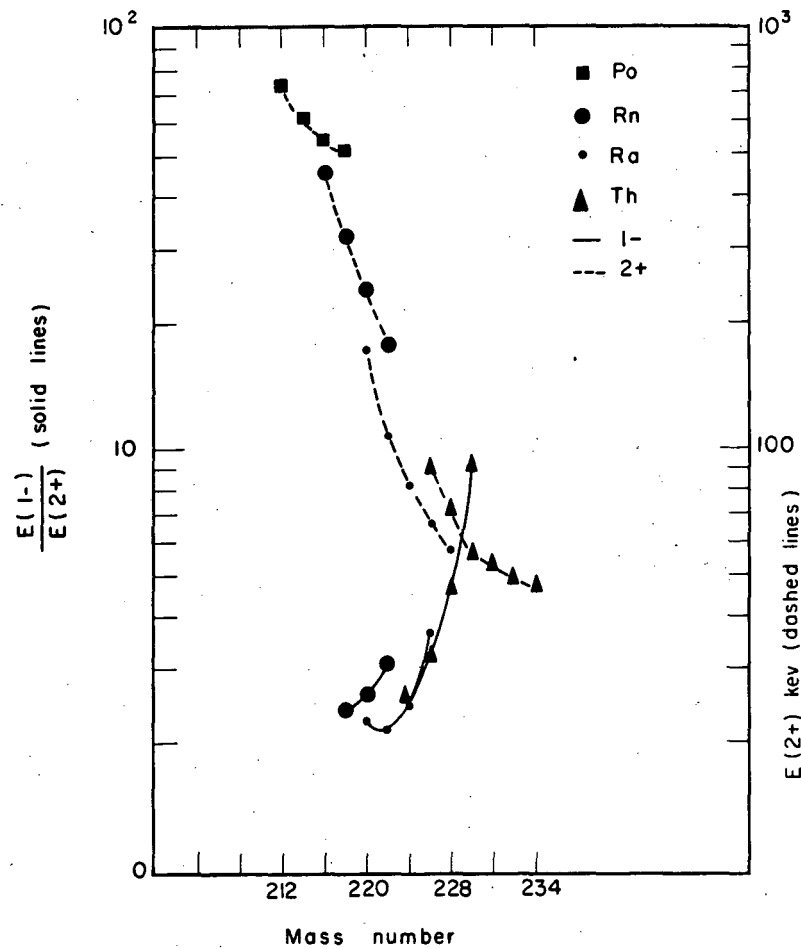


Fig. 42. Even parity states in even-even nuclides.



MU-23336

Fig. 43. Even and odd parity states in even-even nuclides.

interference from the  $U^{229}$  family so no gamma ray could be seen. That this is the first excited state seems quite obvious for the following reasons: (a) its hindrance factor is 1 which is characteristic of that of other first excited states in this region,<sup>41</sup> (b) its energy agrees with that predicted from the trend established by other first excited states in thorium isotopes (Figure 43). As the first excited state its spin and parity are almost certainly 2+.

The 152 and 246 kev gamma rays (Section 2b) indicate the existence of a level at 246 kev. The energy of the 152 kev gamma ray is in good agreement with the energy difference between the 246 and 93 kev levels, and so it is interpreted to be the de-excitation of the 246 kev level to the 93 kev level. Since de-excitation is observed to states of spin 0 and 2, the spin of the 246 kev level must be 1 or 2. From a consideration of the states observed in other even-even nuclei in this region and the fact that alpha decay from a parent of spin and parity 0+ populates this state, it must have spin and parity 1- or 2+. If the state is 2'+, then in terms of the asymmetric rotor model one can calculate a value of  $\gamma = 24^0$  from the energy ratio of the 2'+ state to the 2+. The ratio of the reduced transition probability is about  $\frac{B(E2; 2' \rightarrow 2+)}{B(E2; 2' \rightarrow 0+)} \approx 15$ . Assuming the transition  $2' \rightarrow 2+$  is all E2,<sup>48</sup> the experimental ratio is  $9 \pm 4$ . This is an upper limit for if there is any M1 admixture this quantity would be less. If this assignment were correct, then the most probable assignment for the level at 280 kev would be 4+. From the energy of this level and that of the 2+, a value of  $22^0$  was calculated; with vibration-rotation interaction this value would be even lower. This value is lower than the value of  $24^0$  which is opposite to that observed in the comparison of the experimental data with theory, i.e.,  $\gamma$  values calculated from ratios of 4+ to 2+ should be higher than those calculated from 2'+ and 2+.<sup>48</sup> This does not make the 2+ assignment seem very reasonable.

If the level were 1- then the ratio of the reduced transition probabilities would be  $\frac{B(E1; 1- \rightarrow 0+)}{B(E1; 1- \rightarrow 2+)} = 0.5$ . The experimental ratio is  $0.4 \pm 0.2$ . The energy of this level is in good agreement with the trend established by the energies of the 1- levels of the other thorium isotopes (Figure 43).

From a comparison of the reduced transition probability ratios the 2+ assignment cannot be definitely ruled out. However a 1- assignment seems much more compatible with theoretical arguments and the experimental data on other nuclides in this region.

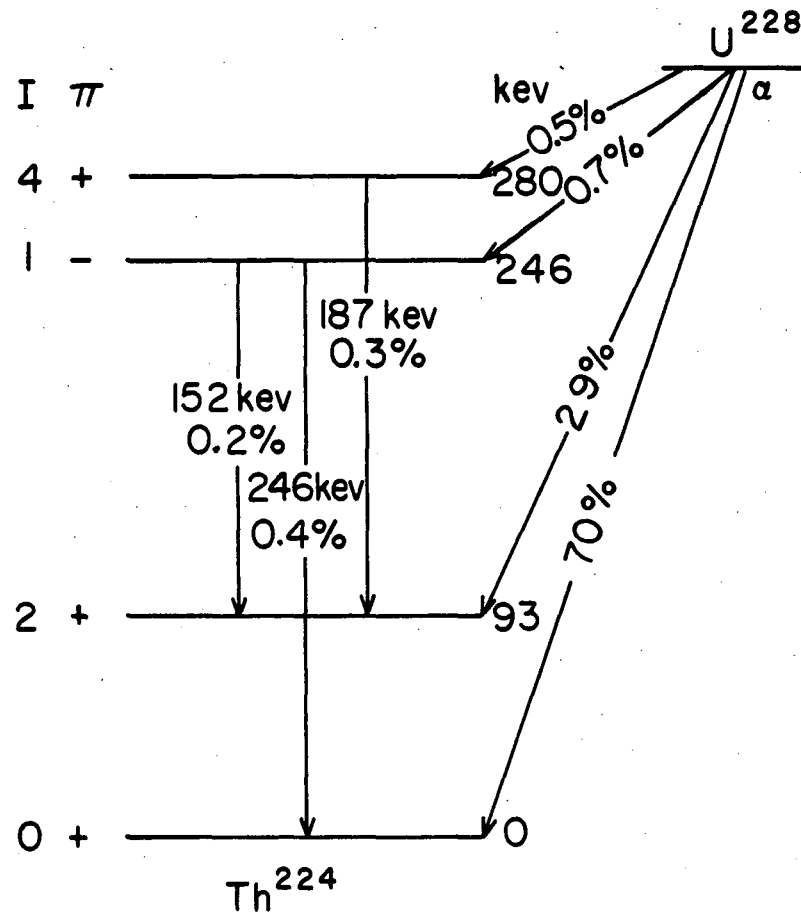
The 187 kev gamma must de-excite a higher lying level to the 93 kev state (the level cannot be at 187 kev because it would not have been included in the alpha gate) which indicates the existence of a state at 280 kev. Considering the levels observed in other even-even nuclides in this region, the only reasonable assignments are 2'+ and 4+. Assuming the state were 2'+, a  $\gamma$  value of 22.5° was calculated indicating the reduced E2 transition probability ratio to be about 9. Thus, one would expect a crossover transition of about the same intensity as the cascade. The fact that none was observed does not rule out the 2'+ assignment. A gamma ray of this intensity and energy would probably not have been detected because of interference from Th<sup>225</sup> radiation.

The energy ratio of this state to the 2+ state is 3.01. If one considers the level to be 4+, a glance at Figure 42 shows this is in excellent agreement with what would be expected (this ratio in Ra<sup>224</sup> is 3.02). It is possible that this agreement is fortuitous in the sense that the 2'+ level could drop down to this value at mass number 224 from 17 in Th<sup>228</sup>. However, if this were the case, a 2'+ level should probably have been observed in Ra<sup>224</sup> in the same energy region which it was not. Therefore the existence of a 2'+ level in Th<sup>224</sup> at this energy is not very probable; therefore the 4+ assignment is much preferred.

The decay scheme summarizing the data is presented in Figure 44. Since alpha groups responsible for the population of the two uppermost levels were not seen directly, and their existence inferred from gamma ray data, their existence is not completely certain. They are indicated by a broken line.

## (2) Thorium-224:

There can be little doubt about the character of the 177 kev level in Ra<sup>220</sup>. The value of the hindrance factor of 1 and observation of E2 radiation de-exciting the level indicate an unambiguous assignment of 2+. This is in agreement with the character of other first excited



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Fig. 44. Decay scheme of  $U^{228}$ .

states in even-even nuclei. Referring to Figure 43 it is seen that the energy is in agreement with what is obtained from an extrapolation of the energy curve of the 2+ states in the other radium isotopes.

An alpha group was observed in the gamma-alpha coincidence spectra at 6.73 Mev (Section 2 a-4) and when gating on this group gamma rays of 145, 177, 235, 295, and 410 kev were observed. The 410 kev gamma must de-excite a level at 410 kev to the ground state to have been seen in the alpha gate. The difference in energy between this and the 177 kev level is 233 kev which agrees quite closely with the energy of the 235 kev gamma ray. This gamma ray is thus taken as de-exciting the 410 kev level to the 2+ state.

Using arguments similar to those used for the state at 246 kev in the  $U^{228}$  decay (previous section) this state can be either 2'+ or 1-. With the assumption that the state is 2'+ the value calculated for  $\gamma$  was  $26^\circ$  and the reduced E2 transition probability ratio of cascade to crossover, calculated from the asymmetric rotor theory was about 30. Assuming the cascade transition was  $E2^{47}$  the experimental value was  $8 \pm 4$ . The only reasonable assignment for the 475 kev level would then be 4+ (1- is not possible since no 475 kev radiation is observed). In this case the value calculated would be  $29^\circ$  (without vibration-rotation interaction correction) compared with  $26^\circ$ . This at least does not conflict with the agreement of experiment and theory in other cases.<sup>43,47,48</sup> If this assignment is correct then its apparent absence in  $Ra^{222}$  must be explained.<sup>53</sup> According to the trends observed experimentally and also according to theory, as nuclides get farther from closed shells the ratio  $R_2$  increases (Figure 42). Therefore one would expect that the 2'+ state would be at an energy ratio higher than that in  $Ra^{220}$ , i.e., greater than 255 kev. The only level which has been seen above this energy is at 310 kev with a tentative 4+ assignment. This is not a definite assignment but very probable. Let us assume therefore, that this is really a 2'+ state. The  $\gamma$  value would be  $23^\circ$ ; the reduced E2 transition probability for cascade to crossover would be about 10. This indicates that the 310 kev transition is about the same intensity as that of the 200 kev transition which would be about 0.6 percent (this is assuming the cascade transition is mainly E2).<sup>48</sup>

This intensity together with that of the cascade transition would require an alpha population of over 1 percent. The experimental value is 0.6 percent. This tends to rule out a  $2^+_{1/2}$  assignment for this state. Also, a transition of this intensity to the ground state probably would have been detected (difficulty arose from interference from the 325 kev gamma ray in the decay of  $\text{Ra}^{222}$ ). This does not rule out the possibility of two unresolved levels at this energy, a  $4^+$  and  $2^+_{1/2}$ . Since the total alpha population is 0.6 percent to these states the alpha population to the  $2^+_{1/2}$  state would have to be less than 0.1 percent. The hindrance factor would then be greater than 30. This is to be compared with a hindrance factor of 2 for the 410 kev state in  $\text{Ra}^{220}$ . In view of this large difference a  $2^+_{1/2}$  assignment does not seem at all reasonable for this state.

In the same set of experiments with  $\text{Ra}^{222}$  a gamma-gamma coincidence experiment was performed in which the 111 kev gamma ray was used as the gating radiation.<sup>53</sup> No gamma radiation higher in energy than 200 kev was observed. (Higher energy radiation in coincidence would have been detected but its energy would not have been known.) Assuming a hindrance factor of 5 and considering their data and the characteristics of de-excitation from a  $2^+_{1/2}$  state, the following statement can be made: there is no  $2^+_{1/2}$  state, within 700 kev of the ground state in  $\text{Ra}^{222}$ . Of course if the alpha hindrance factor were much greater this energy limit would be lower, but then the character of the state would be expected to be different from that of the 410 kev level in  $\text{Ra}^{220}$ .

Assuming a  $1^-$  character for the 410 kev level then the experimental reduced E1 transition probability ratio of cascade to crossover is  $0.4 \pm 0.2$  compared with 0.5 for other nuclides. Further, a  $1^-$  state is certainly expected in this mass region (Figure 43). On the basis of the foregoing arguments, the assignment of the state is made as  $1^-$ .

The 295 kev radiation must de-excite a level at 475 kev to be in coincidence in the gating alpha group of 6.73 Mev. Considering the character of other levels in this region the only reasonable assignments are  $2^+_{1/2}$  and  $4^+$ . Proceeding in the analysis in the same way the  $\gamma$  value calculated for this nucleus assuming the state to be  $2^+_{1/2}$  is  $24^0$ .

The reduced E2 transition probability ratio of cascade to crossover is 15. Therefore the experimental gamma ray cascade and crossover intensities should be approximately equal. No crossover radiation was observed. This coupled with the argument of the  $4+$  level in  $\text{Ra}^{222}$  rules out a  $2^+$  assignment. An assignment of  $4+$  is then made for the 475 kev level in  $\text{Ra}^{220}$ . The alpha hindrance factor of 3 certainly does not conflict with this and it is seen in Figure 42 that this assignment is in very good agreement with the observation of  $4+$  levels in other nuclides in this region.\*

The 88 and 177 kev radiation observed when gating on the 6.73 Mev complex alpha group result as a de-excitation of the 410 and 475 kev levels through the 177 kev level. The relatively high K x-ray intensity is probably due to failure to subtract out enough of the contribution from the de-excitation of levels populated in the decay of  $\text{Ra}^{221}$  which has relatively intense radiation of this energy. The intensity of the 177 kev gamma ray is about 50 percent too high if it is populated only by the 235 and 295 kev transition. One possible explanation is the failure to subtract enough of the chance coincidence counts resulting from a fairly intense gamma ray of about this energy.

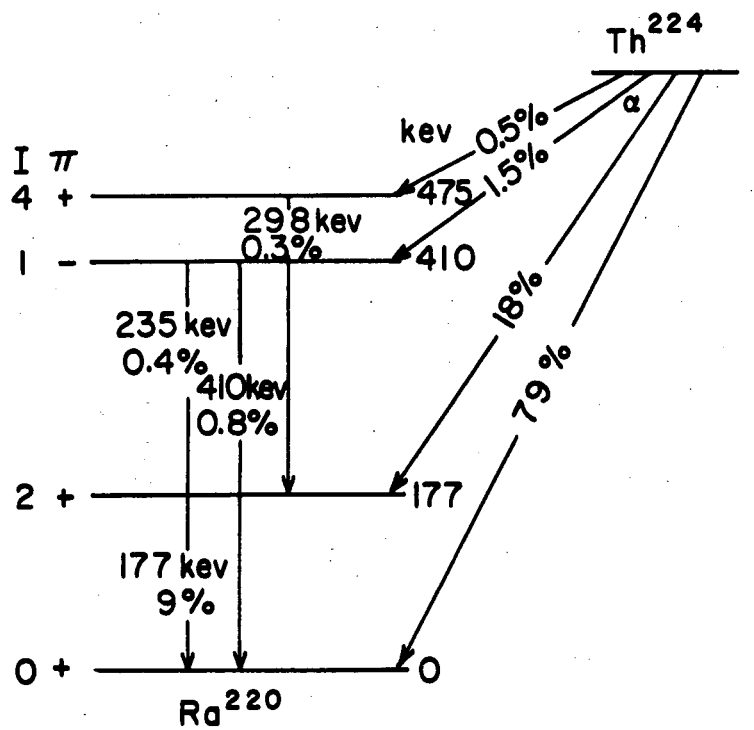
The existence of the 145 kev gamma ray is somewhat disturbing. Its existence is attributed to scattered radiation since its intensity was observed to fluctuate with respect to that of the other gamma rays for a given set of experiments. The decay scheme is shown in Figure 45.

### (3) Radium-220:

It has been shown (Section 2, a-2) that the 465 kev gamma ray is associated with the decay of  $\text{Ra}^{220}$  and indicates a level in  $\text{Rn}^{216}$  465 kev above the ground state. It is not possible to determine the multipolarity of the transition from the data; however, the correct assignment of the state is probably  $2+$ . With reference to Figure 46, from an extrapolation of the energies of the  $2+$  states in other radon isotopes a  $2+$  state of about this energy would be expected in  $\text{Rn}^{216}$ .

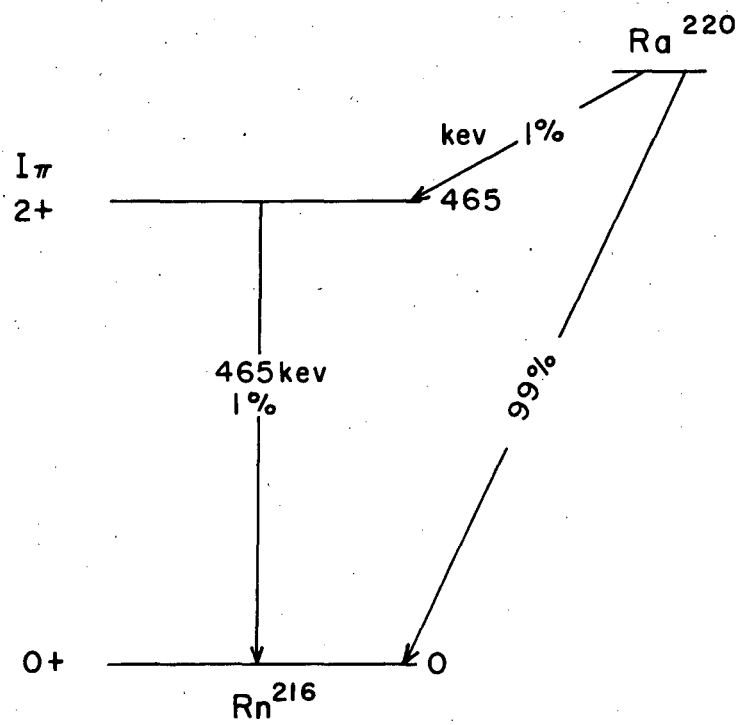
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\* It might be added that this also lends support to the assignment of  $4+$  for the 850 kev level in  $\text{Rn}^{218}$  in reference 49.



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Fig. 45. Decay scheme of  $\text{Th}^{224}$ .



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Fig. 46. Decay scheme of  $\text{Ra}^{220}$ .

Since there is no evidence for the existence of lower lying excited states, then this is probably the first excited state in which case a  $2+$  assignment would be expected. The decay scheme is shown in Figure 46.

(4) Radon-216:

Only the ground state alpha group was observed (Table XI). No gamma rays were observed which could be attributed to the decay of  $\text{Rn}^{216}$ . From the beta decay of  $\text{Bi}^{212}$  a level at 729 kev is populated in  $\text{Po}^{212}$  and assigned the spin and parity  $2+$ . The limit of detection of this level populated by alpha decay in the present experiments was 0.2 percent; thus the alpha hindrance factor is greater than 3. The hindrance factors for alpha decay to the  $2+$  level in other Po isotopes are 2.1, 1.3, and 3.1 respectively, for  $\text{Rn}^{222}$ ,  $\text{Rn}^{220}$ , and  $\text{Rn}^{218}$  decay.<sup>41</sup> Therefore the hindrance factor in the case of  $\text{Rn}^{216}$  decay is probably not much greater than 3.

c. Discussion

It has been suggested that sharp discontinuities occur in the character of the levels between the spherical and non-spherical regions; specifically, in the heavy element region between proton numbers 86 and 88<sup>55</sup> and neutron number 134 to 136.<sup>56</sup> If the assignments of the  $4+$  states are correct, Figure 42 shows a gradual change in the ratio  $R_4$  through this region of transition with no abrupt changes in the character of this state.\* This is similar to the region of transition below the closed shell at  $\text{Pb}^{208}$  where the ratio  $R_4$  changes gradually from 3.33 in Hf to 2.42 in Hg,\*\* Figure 42. This similarity between the two transition regions seems to vanish when the  $2'+$  states are compared. In the region of osmium there is a gradual transition in the ratio  $R_2$  with respect to mass number; this gradual transition apparently does not exist in the heavy element region although the data on  $2'+$  states in this region are rather incomplete (Figure 42).

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\* This conclusion was previously reached by F. Stephens et al.<sup>50</sup>

\*\* The gradual transition in this region was suggested by Goldhaber.<sup>55</sup>

It would seem that in analogy to the osmium region the  $2^+$  state should have been seen in  $\text{Ra}^{220}$  and  $\text{Ra}^{222}$  at relatively low energies. From the arguments given in Section b, this seems rather remote. The asymmetric rotor theory (without vibration-rotation interaction) would also predict  $2^+$  states in this region. It was stated in Section a that according to the theory for values of  $\gamma > 21.5^\circ$ , the  $2^+$  level should lie below the  $4^+$  levels. Figure 47 shows  $\gamma$  plotted as a function of  $R_4$ . The uppermost line was determined from theory. The intermediate line was drawn through the points for which the value of gamma had been determined from the ratio  $R_2$  in Equation 7. It is essentially a vibration-rotation interaction correction. Now in  $\text{Os}^{190}$  ( $\gamma = 22^\circ$ ) the  $2^+$  level is very close to the  $4^+$  level (9 kev). In  $\text{Os}^{188}$  ( $\gamma = 19^\circ$ ) the  $2^+$  level is 155 kev above the  $4^+$  and in  $\text{Os}^{192}$  it is 91 kev below it. Clearly then the experimental data and theory are in good agreement.

Since the levels of the  $2^+$  states are not known over much of the trans-lead region, the values of gamma were computed using the  $R_4$  ratios and the corrections applied that were used in the osmium region. (In the case of  $\text{Th}^{228}$ ,  $\text{Th}^{230}$ ,  $\text{U}^{232}$ ,  $\text{U}^{234}$ ,  $\text{Pu}^{238}$ , and  $\text{Pu}^{240}$ , the gamma values were computed making use of their  $2^+$  states which are plotted in Figure 47). It became obvious that  $\text{Ra}^{222}$  ( $\gamma = 25^\circ$ ) and  $\text{Ra}^{220}$  ( $\gamma = 29^\circ$ ) should have their  $2^+$  state below their  $4^+$ . In fact, according to this analysis  $\text{Ra}^{220}$  should be almost spherical. If the  $4^+$  assignments in  $\text{Ra}^{220}$  and  $\text{Ra}^{222}$  are correct then the theory with a small vibration-rotation interaction ( $\mu = 0.25$ ) does not explain the data, and a substantially larger value of  $\mu$  is needed.

It has already been mentioned that a sharp transition occurs between proton numbers 86 and 88. There is also one that occurs between neutron numbers 88 and 90.<sup>55</sup> It was also noted (Section a) that the theory (without vibration-rotation interaction) cannot explain the spectra of  $\text{Sm}^{152}$ ,  $\text{Gd}^{154}$ , and  $\text{Gd}^{156}$  nuclides in this other transition region. The  $2^+$  levels have been observed for them which enabled the calculation of a gamma value. These points are also plotted in Figure 47. A very large deviation is noted between the gamma values computed using  $R_2$  and those using  $R_4$ . In other words

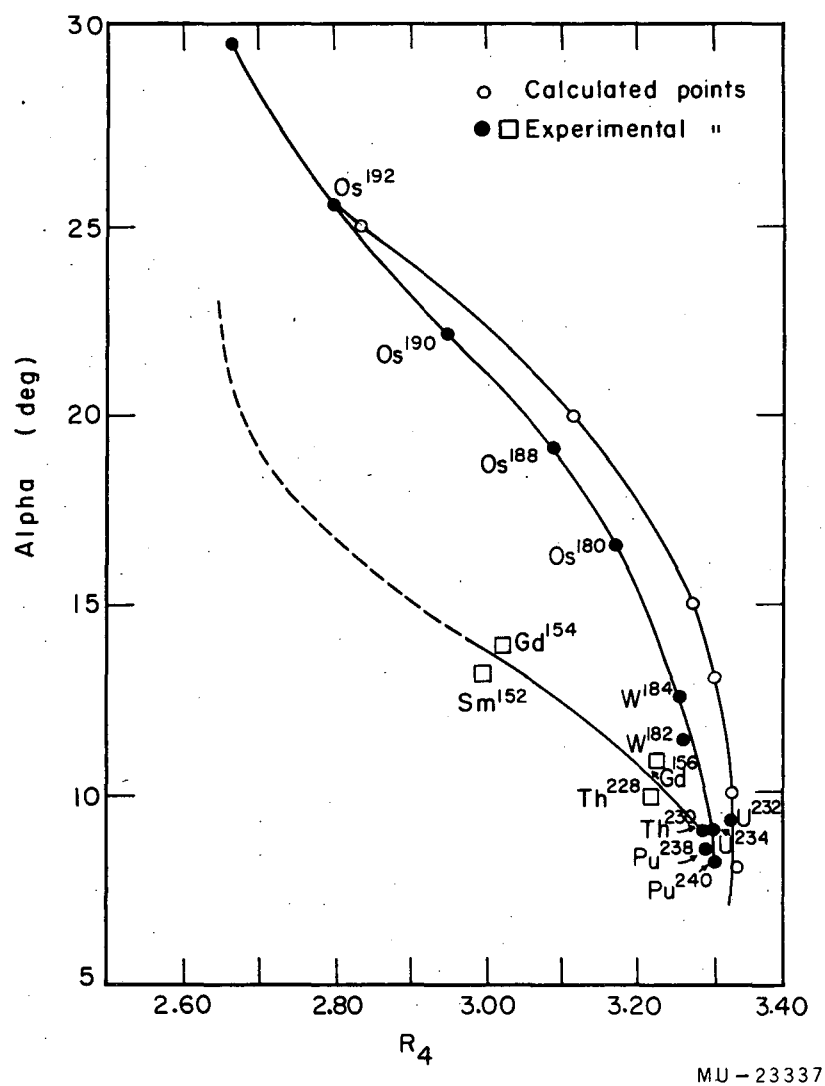


Fig. 47. Diagram showing variation of  $\gamma$  with  $R_4$ . Uppermost curve shows theoretical prediction; lower two curves are drawn through experimental points.

$\mu$  is substantially greater here ( $\mu \sim 0.36$ ) than in the osmium region ( $\mu \sim 0.25$ ). It is also worthy of note that  $\text{Th}^{228}$  ( $\mu = 0.27$ ) on the fringe of the heavy element transition region has almost the same gamma value as  $\text{Gd}^{156}$  ( $\mu = 0.26$ ). If a line is drawn through these four points and extrapolated in some manner to lower values of  $R_4$ , it is seen that  $\text{Ra}^{220}$  and  $\text{Ra}^{222}$  can be made to fall below  $22^\circ$  by placing them on the curve, i.e., the value of  $\mu$  is such that  $\gamma$  is less than  $22^\circ$ . It is interesting to see that from this plot there appears to be a marked similarity between the Sm and Ra regions as opposed to the osmium region. This of course is reflected in the value of  $\mu$ .

For the samarium region  $\mu$  is about 0.37; for the radium region  $\mu$  must be greater than 0.4, while for the osmium region it is about 0.25. This tends to indicate that the nuclei in the samarium and radium regions are much "softer" to deformation than those in the osmium region. This is consistent with the fact that 1- states, which are attributed to an unsymmetrical deformation of the nucleus (octopole vibrations), are observed to be quite low in the radium region but not in the osmium region. It is interesting to note also that the value of  $\mu$  for  $\text{Rn}^{218}$  is about 0.27. It appears therefore that the value of  $\mu$  must pass through a maximum value. It would be interesting to see if this maximum occurred at the same mass number as the energy minimum for the 1- states (Figure 43).

In conclusion then, it may be stated that although the  $R_4$  ratio seems to undergo a gradual variation with mass number in the region of radium, the  $R_2$  ratio seems to exhibit a sudden jump between proton numbers 86 and 88. This latter statement is concluded from the absence of  $2^+$  states and must be regarded as tentative.

It appears that the theory with the vibration-rotation interaction correction can explain the apparently high lying  $2^+$  states on the basis of the tendency of the nuclei in the region to readily undergo deformation. This explains why the radium region is much more like the samarium region than the osmium region. However, the problem of why the nuclei in the radium and samarium regions are "softer" than those in the osmium region remains to be solved. The existence of the 88-90 neutron numbers and 86-88 proton numbers may provide the answer.

The data on the 1- states are in agreement with the previous data obtained on other even-even nuclei. In Figure 46 there appears to be a

gap between the radon points and those of radium and thorium. In view of the above discussion, it is tempting to attribute this to the "critical" proton numbers 86 and 88; however, this apparent relationship may only be accidental.

## C. Other Studies

### 1. Uranium-233

$U^{233}$  has been the subject of several investigations. Studier<sup>57</sup> observed gamma rays of 40 and 80 kev and their corresponding conversion electrons, as well as a 0.31 Mev gamma ray having an intensity of 0.1 percent. Martin<sup>58</sup> found 39 and 85 kev gamma rays in an intensity of 1 percent of the L x-rays. Prohaska<sup>59</sup> saw gamma rays of 39, 88, and 360 kev in coincidence with 0.2, 0.15, and 0.19 percent respectively, of the alpha particles. He also found conversion electrons having energies 36, 68, and 260 kev in coincidence with 6.2, 0.69, and 0.28 percent respectively, of the alpha particle. Bisgard<sup>60</sup> found electrons corresponding to 43, 56, and 99 kev gamma rays in coincidence with alpha particles. Donlavey<sup>61</sup> observed electrons corresponding to 40 and 90 kev gamma rays in coincidence with 9 and 0.4 percent respectively, of the alpha particles. West, et al.<sup>62</sup> reported L x-rays of thorium, and gamma rays of  $42.8 \pm 0.4$  kev and  $56.1 \pm 0.4$  kev in intensities of  $4 \pm 1 \times 10^{-2}$ ,  $5 \times 10^{-4}$  and  $1 \times 10^{-4}$  per alpha particle. It was later reported<sup>63</sup> that in this latter case the relative intensities were correct but the absolute intensities were too small. Finally, Albouy<sup>64</sup> saw gamma rays of 40 and 90 kev with an L/M ratio of  $3 \pm 1$ .

Asaro<sup>65</sup> observed three groups in the alpha spectrum having intensities 83, 15, and 2 percent. The latter two being 42 and 96 kev respectively, lower in decay energy than the ground state. He interpreted the 80 - 100 and 56 kev radiation as de-exciting the 96 kev level, and the 42.8 kev transition as de-exciting the level at 42 kev. Besides these alpha groups, which Goldin, et al.<sup>66</sup> also observed having relative intensities of 83.5,  $14.9 \pm 0.7$ , and  $1.6 \pm 0.2$  percent, the latter two having energies  $42.5 \pm 0.4$  and  $98.3 \pm 1$  kev less than the  $\alpha_0$  group, they saw a new group populating a level  $333 \pm 5$  kev above the ground state in an abundance of  $0.03 \pm 0.01$  percent. They interpreted the 300 - 360 kev radiation as de-exciting this level to the ground state and first two excited states. Two more alpha groups were later reported by them<sup>67</sup> at energies of 161 and 234 kev less than  $\alpha_0$  having intensities of  $0.07 \pm 0.04$  and  $0.04 \pm 0.02$  percent respectively (Table XIII).

Experimentally, the ratios of the energies of the levels were 1: 2.3 : 3.8 : 5.5 : 7.7; theoretically these ratios calculated from Equation 2

Table XIII. Alpha Groups of  $U^{233}$

| Goldin <u>et. al.</u> <sup>67</sup>        |                  | Dzhelepov <u>et. al.</u> <sup>68</sup>     |                  |
|--------------------------------------------|------------------|--------------------------------------------|------------------|
| Excited State<br>Energy (kev) <sup>a</sup> | Abundance<br>(%) | Excited State<br>Energy (kev) <sup>a</sup> | Abundance<br>(%) |
| 0                                          | 83.5             | 0                                          | 83.4             |
| 43.2 ± 0.4                                 | 14.9 ± 0.7       | 43                                         | 14.9             |
| 100 ± 1                                    | 1.6 ± 0.2        | 100                                        | 1.6              |
| 164                                        | 0.07 ± 0.04      | 166                                        | 0.060 ± 0.015    |
| 237                                        | 0.04 ± 0.02      | 234                                        | <0.007           |
| 333 ± 5                                    | 0.03 ± 0.01      | 316                                        | 0.033 ± 0.008    |
|                                            |                  | 364                                        | <0.005           |
| Tret'yakov <u>et. al.</u> <sup>69</sup>    |                  | This Work                                  |                  |
| 0 <sup>b</sup>                             | 83.5             | 0                                          | 83.7             |
| 29 <sup>c</sup>                            | 0.53             | 43 ± 1                                     | 14.1 ± 0.7       |
| 42 <sup>b</sup>                            | 14.9             | 99 ± 2                                     | 1.9 ± 0.3        |
| 71                                         | 0.3              | 166 ± 4                                    | 0.05 ± 0.03      |
| 97 <sup>b</sup>                            | 1.6              | 237                                        | <0.02            |
| 131 <sup>c</sup>                           | 0.05             | 320 ± 4                                    | 0.04 ± 0.02      |
| 164 <sup>b</sup>                           | 0.07             |                                            |                  |
| 320 <sup>b</sup>                           | 0.03             |                                            |                  |

<sup>a</sup> All energies measured relative to ground state energy taken as 4.816 Mev.

<sup>b</sup> The data for these levels was taken from reference 67.

<sup>c</sup> The existence of these groups was questionable in the alpha spectrum.

were 1 : 2.28 : 3.89 : 5.72 : 7.85. Because of this agreement they concluded all the observed levels were members of a rotational band based on spin  $5/2$  for the ground state. They noted that although the rapid and regular variation of the intensity observed for the first four levels could be described by several theoretical formulas, none was capable of describing the extremely slow variation in intensity observed for the last three members. They suggested that possibly the departures from the theoretical formulas were inevitable for states of high angular momenta and particularly important for  $U^{233}$ . The data are summarized in Table XIII.

Because of this apparent conflict between experiment and theory and also uncertainty in the assignment of the level at 333 kev as  $15/2$ , since gamma radiation of about this energy had been observed, and investigation of  $U^{233}$  was undertaken.

It was not until this study had been completed that two new papers appeared on the alpha decay of  $U^{233}$ . Dzhelepov et al.<sup>68</sup> studied the alpha spectrum and Tretyakov et al.<sup>69</sup> studied the alpha spectrum and internal conversion electron spectrum. The results are shown in Tables XIII and XIV. The data obtained in the present study will be presented and discussed in the light of this recent information.

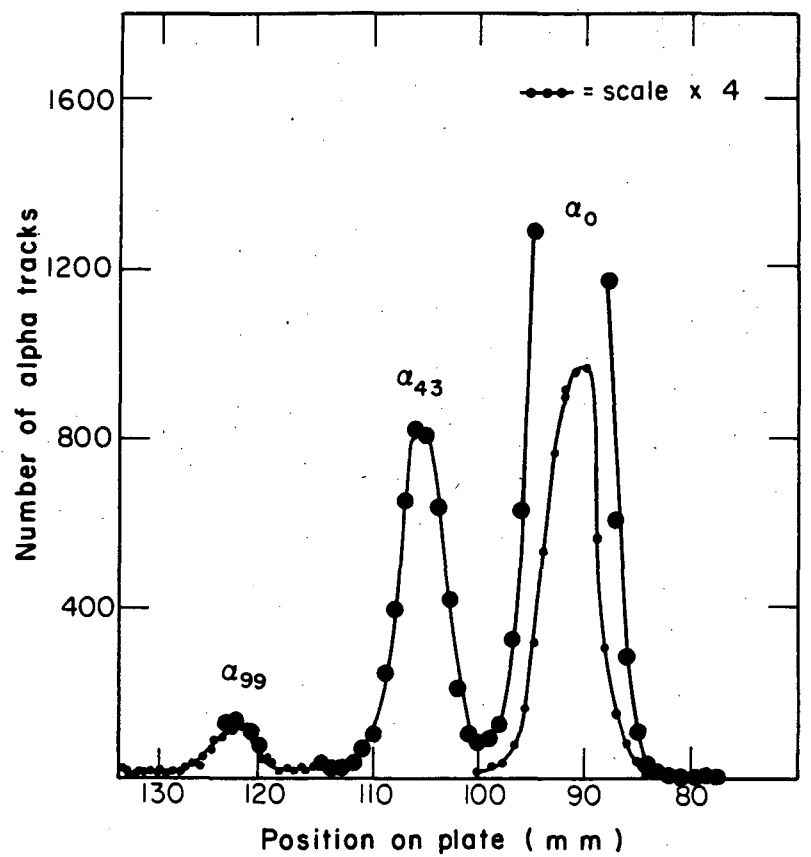
#### a. Alpha Spectrum

Several runs were made with samples purified immediately before each run by passing the uranium through an anion exchange column. Sources of about  $2 \times 10^5$  alpha disintegrations per minute were used; more active sources provided too much mass and therefore contributed to low energy tailing which completely masked the groups of lower energy and low intensity. The length of the exposures varied from two days to two weeks. Figure 48 is a spectrum of the three most intense groups of  $U^{233}$  obtained from a 2-day exposure; Figure 49 shows that region of the spectrum exhibiting the low intensity groups obtained with a 10-day exposure. Table XIII shows that the data are in agreement with that of Dzhelepov and Tret'yakov. The 237 kev state of Golden et al. is seen to be in much lower abundance. The fact that  $\alpha_{29}$ ,  $\alpha_{71}$ , and  $\alpha_{131}$  were not observed in any of the spectra except those of Tret'yakov does not conflict with their data since these groups would not have been observed in the other spectra.

Table XIV. Internal Transitions in  $\text{Th}^{229}$

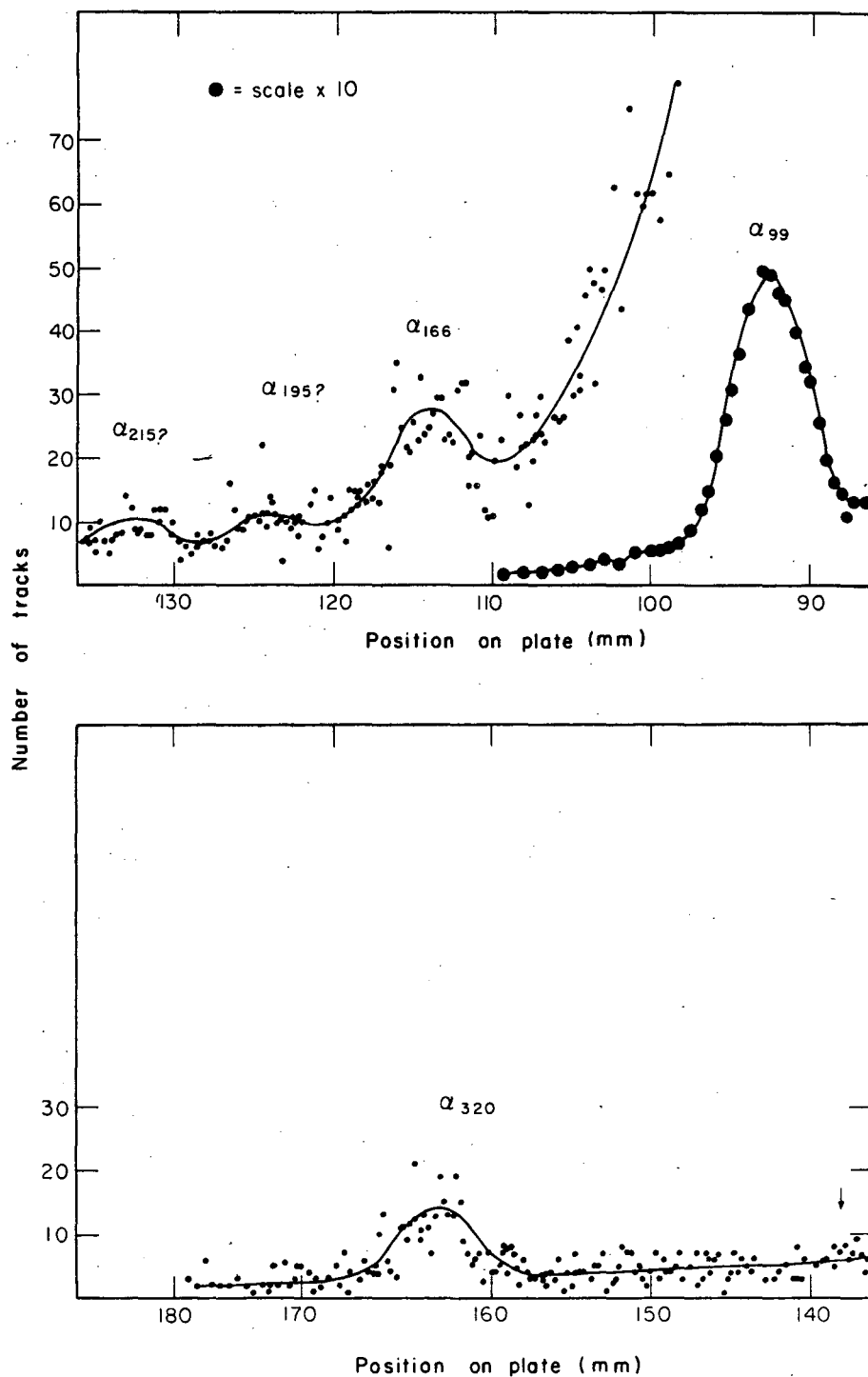
| From Internal Conversion Electron Spectra<br>(Tret'yakov et al.) <sup>13</sup> |                                                         |                    | From Gamma Ray<br>Spectra-This Work |                                           |
|--------------------------------------------------------------------------------|---------------------------------------------------------|--------------------|-------------------------------------|-------------------------------------------|
| Transition<br>Energy (kev)                                                     | Transition<br>Intensity<br>Transitions<br>$10^2$ alphas | Multi-<br>polarity | Energy<br>(kev)                     | Intensity<br>Transitions<br>$10^2$ alphas |
| 29.1 $\pm$ 0.2                                                                 | 0.7 $\pm$ 0.4                                           | M1 <sup>a</sup>    |                                     |                                           |
| 42.4 $\pm$ 0.2                                                                 | 16.0                                                    | 80% M1             | 42 $\pm$ 2                          | 0.065 $\pm$ 0.010                         |
| 54.4 $\pm$ 0.5                                                                 | 1.0 $\pm$ 0.3                                           | 20% E2<br>M1 + E2  | 57 $\pm$ 4                          | 0.018 $\pm$ 0.004                         |
| 66.0 $\pm$ 1.0                                                                 | 0.05                                                    |                    |                                     |                                           |
| 71.4 $\pm$ 0.6                                                                 | 0.30                                                    |                    | 72                                  | 0.011                                     |
| 97.3 $\pm$ 0.3                                                                 | 0.6                                                     | E2                 | 97 $\pm$ 2                          | 0.041 $\pm$ 0.006                         |
| 103.0 $\pm$ 1.0                                                                | 0.01                                                    |                    |                                     |                                           |
| 121.0 $\pm$ 0.3                                                                | 0.03                                                    | E2                 | 118                                 | 0.012                                     |
|                                                                                |                                                         |                    | 142                                 | 0.008                                     |
|                                                                                |                                                         |                    | 162                                 | 0.006                                     |
|                                                                                |                                                         |                    | 182                                 | 0.001                                     |
|                                                                                |                                                         |                    | 211                                 | 0.006                                     |
| 245.3 $\pm$ 0.5                                                                | 0.015                                                   | M1                 | 242                                 | 0.005                                     |
| 248.6 $\pm$ 0.8                                                                | 0.008                                                   |                    |                                     |                                           |
| 277.8 $\pm$ 1.5                                                                | 0.005                                                   | M1 <sup>a</sup>    | 282                                 | 0.007                                     |
| 291.5 $\pm$ 0.5                                                                | 0.012                                                   | M1                 |                                     |                                           |
| 317.0 $\pm$ 1.5                                                                | 0.02                                                    | M1                 | 318                                 | 0.01                                      |
| 321.0 $\pm$ 1.5                                                                | 0.008                                                   | M1 <sup>a</sup>    |                                     |                                           |
| 366.0 $\pm$ 2.0                                                                | 0.005                                                   | M1 <sup>a</sup>    | 370                                 | 0.001                                     |

<sup>a</sup> Assignment uncertain.



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Fig. 48. Alpha particle spectrum of high intensity groups of  $U^{233}$ .



MUB-641

Fig. 49. Alpha particle spectrum of low intensity groups of  $U^{233}$ .  
 Arrow shows location where  $\alpha_{237}$  would appear.

There appear to be two new groups,  $\alpha_{195}$  and  $\alpha_{215}$ , but their existence must be considered doubtful because statistical fluctuation in the background could account for the apparent peaks. The intensity of these groups would be less than 0.02 percent.

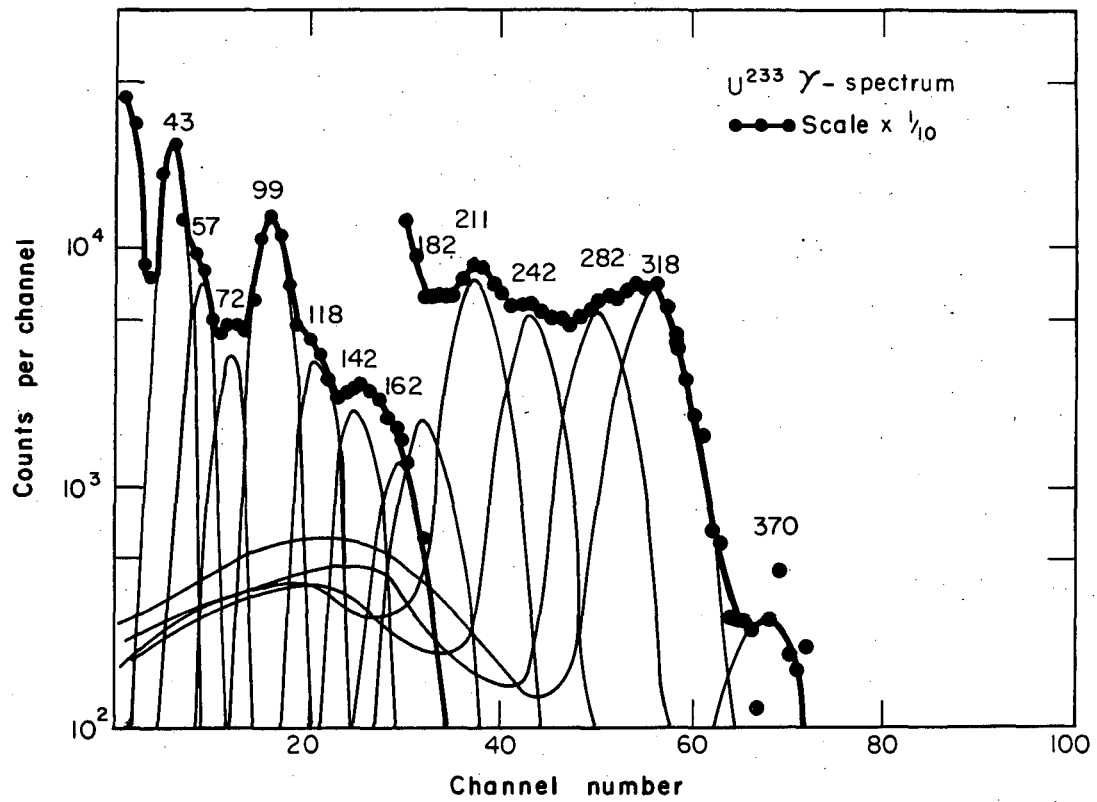
b. Gamma Spectra

The gamma ray singles spectrum obtained from a newly purified sample of  $U^{233}$  evaporated onto a nickel plate is shown in Figure 50. An alpha-gamma coincidence spectrum was also obtained which proved to be exactly the same. The energies and intensities are given in Table XIV and the first two columns of Table XV. The gamma rays were resolved graphically but this process was aided immeasurably by the results of the alpha-gamma, and gamma-gamma coincidence studies. This increased greatly the accuracy of the method. The only other alpha active impurity in the sample was  $U^{232}$  which was 1.6 percent of the counting rate; since the runs were made immediately after chemical purification the daughters of  $U^{232}$  could not have grown into an appreciable extent. Considering the energy and intensity of its gamma rays, then its contribution to the corresponding energy region of the  $U^{233}$  spectrum would be down by at least an order of magnitude from the intensity of the  $U^{233}$  gamma rays. It is therefore certain that the gamma rays observed are associated with the alpha decay of  $U^{233}$ .

c. Coincidence Spectra

(1) Gamma-Gamma Coincidence Spectra:

All energy regions from 42 through 320 kev as well as L x-rays were used as gates so that the gamma spectrum in coincidence with each energy region corresponding to one of the gamma rays listed in Table XV was observed. The sample was sandwiched in between the faces of two NaI (Tl activated) scintillation detectors; the coincidence circuit described in Section I was used, with the coincidence spectra recorded on a 50-channel pulse-height analyzer. When gating on the lower energy regions enough silver or copper adsorber was placed between the sample and the signal detector to substantially reduce radiation scattered from the signal to the gate detector. This reduced in proportion the number of false coincidences.



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Fig. 50. Gamma ray spectrum of U<sup>233</sup>.

The data obtained from these coincidences spectra are summarized in Table XV. The columns (excluding the first two) then represent a gating energy and the intensity of a gamma ray in coincidence with this is recorded opposite the corresponding gamma ray energy of column one. When it was not possible to calculate the intensity, this number was estimated and indicated by a "p" for a partial coincidence (no full coincidences were observed for these cases). In all cases if the coincidence intensity was less than five times smaller than the singles intensity, then it was recorded as no coincidence, indicated by a dash. The 42 kev gamma ray was not recorded in the table because the amount of adsorber used made its intensity in the coincidence runs uncertain. The gamma rays of energy 72, 182, 211, 242, 282, and 320 kev were gated on but had no gamma rays in coincidence with them (L x-rays in coincidence with these would not have been seen).

TABLE XV

| Gamma-Gamma Coincidence Results |                   |                                            |     |     |     |     |     |     |
|---------------------------------|-------------------|--------------------------------------------|-----|-----|-----|-----|-----|-----|
| γ-ray energy (keV)              | Singles intensity | Gate gamma energy (keV)                    |     |     |     |     |     |     |
|                                 |                   | L x-rays                                   | 42  | 57  | 95  | 118 | 142 | 164 |
|                                 |                   | Intensity Photons / 10 <sup>4</sup> Alphas |     |     |     |     |     |     |
| 42                              | 6.5               | -                                          | -   | -   | -   | -   | -   | -   |
| 57                              | 1.8               | 1.9                                        | 1.8 | -   | -   | -   | -   | -   |
| 72                              | 1.1               | 1.3                                        | 1.1 | 0.5 | 0.3 | -   | -   | -   |
| 97                              | 4.1               | 4.1                                        | 3.3 | 1.3 | 0.6 | p   | p   | -   |
| 118                             | 1.2               | 1.2                                        | 0.6 | 0.7 | 0.7 | -   | -   | -   |
| 142                             | 0.8               | 0.6                                        | 0.5 | 0.4 | 0.3 | p   | -   | p   |
| 162                             | 0.6               | 0.4                                        | 0.4 | 0.2 | 0.2 | -   | p   | -   |
| 182                             | 0.1               | 0.2                                        | 0.1 | 0.1 | 0.1 | p   | -   | -   |
| 211                             | 0.6               | 0.3                                        | -   | -   | -   | -   | -   | -   |
| 242                             | 0.5               | 0.6                                        | 0.5 | -   | -   | -   | -   | -   |
| 282                             | 0.7               | 0.4                                        | -   | -   | -   | -   | -   | -   |
| 318                             | 1.0               | -                                          | -   | -   | -   | -   | -   | -   |
| 370                             | 0.1               | -                                          | -   | -   | -   | -   | -   | -   |

## (2) Gamma-Alpha Coincidence:

There was good reason to believe that the alpha spectrum was more complex than thus far observed because of the complex gamma spectrum (Figure 50). In order to see lower intensity alpha groups, a gamma alpha coincidence run was made. All gamma rays of energy greater than 35 kev were used to gate the circuit and the coincident

alpha spectrum detected with the alpha grid chamber. The coincidence circuit described in Section I was used. In this way all the tailing background contribution from  $\alpha_0$  and most of it from  $\alpha_{42}$  and  $\alpha_{97}$  was eliminated; the spectrum obtained from a three-day run is shown in Figure 51. This spectrum confirmed the main alpha groups seen in Figure 48 (with the exception of  $\alpha_0$ ) and in addition showed at least six groups:  $\alpha_{142}$ ,  $\alpha_{210}$ ,  $\alpha_{245}$ ,  $\alpha_{285}$ ,  $\alpha_{318}$ , and  $\alpha_{367}$ . From this spectrum, the existence of  $\alpha_{185}$ , and  $\alpha_{367}$  was not conclusively proven but was certainly suggested. The intensity of the groups can not be calculated directly. It depends on the intensity of the gamma rays de-exciting the levels, and will be estimated in Section 5.

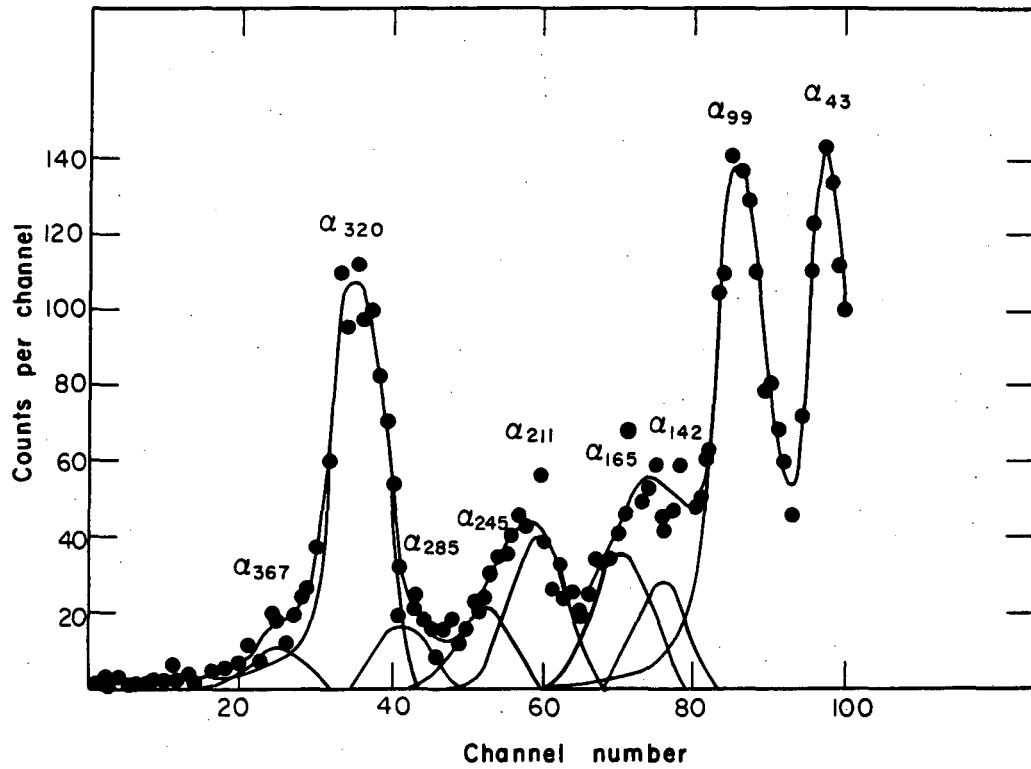
Figure 52 is a gamma-alpha coincidence spectrum gating on L X-rays and gamma rays, using a ring collimator. Besides the relative changes in intensity of the groups, the spectrum is "smeared out" due to the conversion electron effect.

### (3) Alpha-Gamma Coincidence:

With the exception of  $\alpha_{367}$ , all the alpha groups seen in Figure 51 were gated on and the gamma rays observed in coincidence. Since the groups from  $\alpha_{166}$  to  $\alpha_{320}$  were complex, the gate was shifted to include different portions of the peak; a one-day run was made at each setting. These data are recorded in Table XVI where the energy of the gamma ray is given in column 1, and that of the gating alpha given along the row at the top.

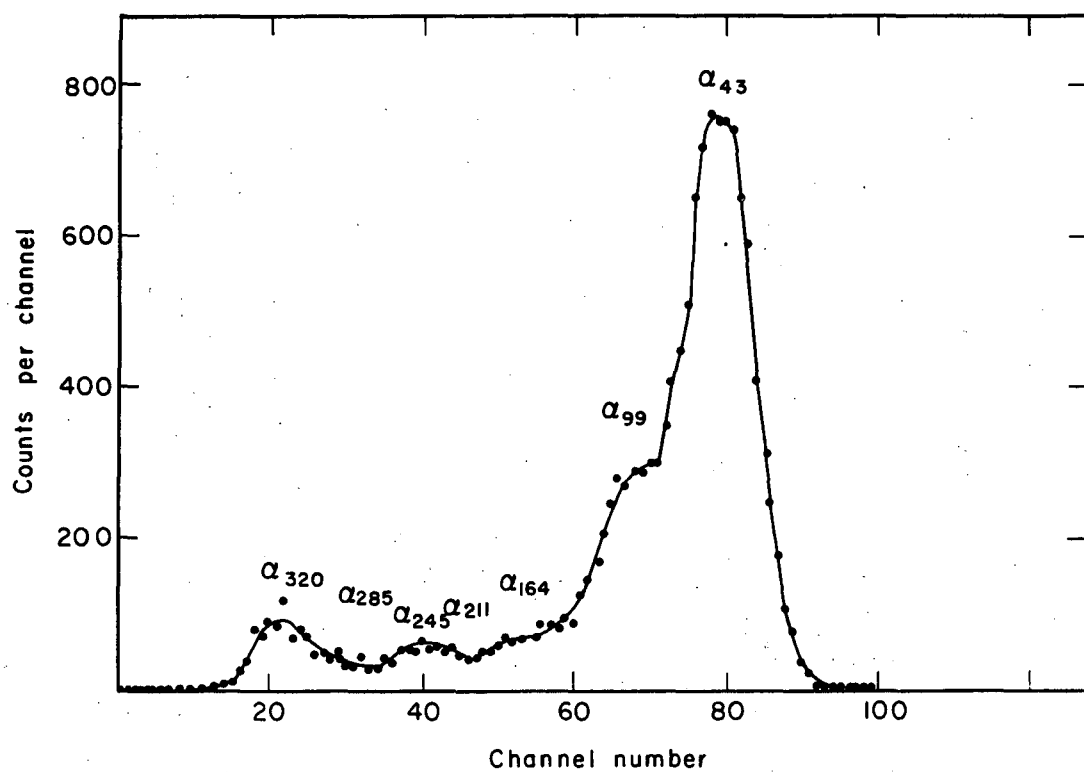
TABLE XVI.

| Alpha-Gamma Coincidence Results   |                    |               |                 |                 |                 |                |                 |                 |                |                 |
|-----------------------------------|--------------------|---------------|-----------------|-----------------|-----------------|----------------|-----------------|-----------------|----------------|-----------------|
| γ ray<br>Energy<br>(kev)          | Gating Alpha Group |               |                 |                 |                 |                |                 |                 |                |                 |
|                                   | $\alpha_{42}$      | $\alpha_{99}$ | $\alpha_{166}-$ | $\alpha_{166}+$ | $\alpha_{211}-$ | $\alpha_{211}$ | $\alpha_{211}+$ | $\alpha_{320}-$ | $\alpha_{320}$ | $\alpha_{320}+$ |
| Intensity Photons / $10^4$ Alphas |                    |               |                 |                 |                 |                |                 |                 |                |                 |
| 42                                | 2                  | 1             | 0.1             | -               | -               | -              | -               | -               | -              | -               |
| 57                                | -                  | .6            | 0.2             | 0.1             | -               | -              | -               | -               | -              | -               |
| 73                                | -                  | -             | 0.2             | 0.1             | 0.1             | -              | -               | -               | 0.05           | 0.05            |
| 99                                | -                  | 1.1           | 0.4             | 0.2             | 0.1             | 0.1            | 0.3             | 0.2             | 0.6            | 0.2             |
| 117                               | -                  | -             | 0.3             | 0.2             | 0.1             | 0.2            | 0.05            | -               | -              | -               |
| 142                               | -                  | -             | 0.3             | 0.2             | 0.2             | 0.1            | -               | 0.05            | 0.05           | -               |
| 162                               | -                  | -             | 0.1             | 0.2             | 0.1             | 0.1            | 0.05            | -               | 0.05           | -               |
| 182                               | -                  | -             | -               | -               | -               | -              | -               | -               | 0.05           | -               |
| 211                               | -                  | -             | -               | -               | 0.2             | 0.2            | -               | 0.1             | -              | -               |
| 242                               | -                  | -             | -               | -               | -               | -              | -               | 0.05            | 0.3            | 0.1             |
| 282                               | -                  | -             | -               | -               | -               | -              | -               | 0.05            | 0.3            | 0.1             |
| 318                               | -                  | -             | -               | -               | -               | -              | -               | 0.1             | 0.4            | 0.2             |



MU-23341

Fig. 51. Gamma ray—alpha particle coincidence spectrum of  $U^{233}$  with 100-count mesh collimator.



MU-23340

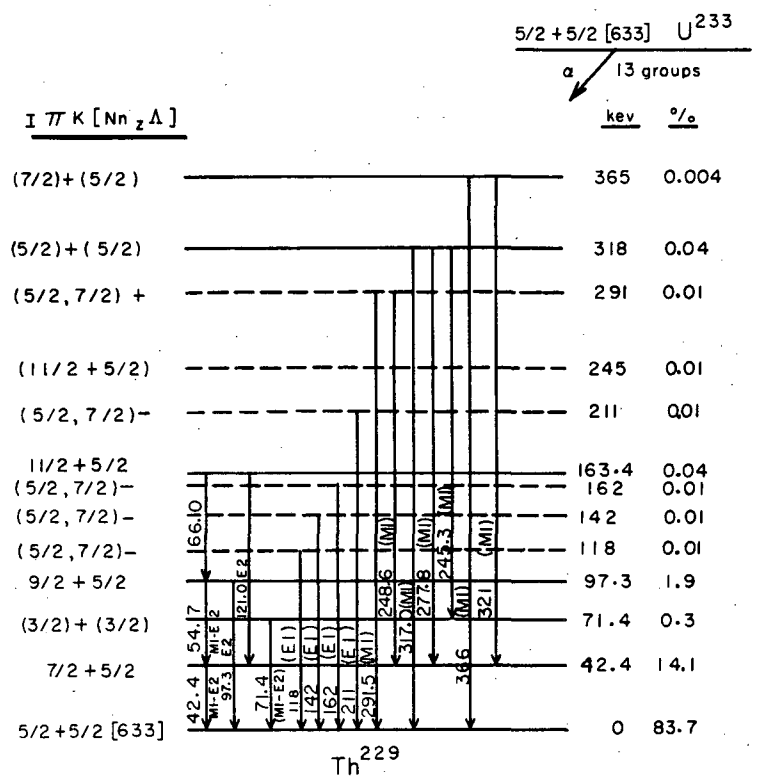
Fig. 52. Gamma ray—alpha particle coincidence spectrum of  $U^{233}$  with ring collimator.

Gating on the low energy side of a group is indicated by a "+" and gating on high energy side by a "-". When no symbol is used the gate was set in the center of the peak. A coincidence is indicated by recording the intensity of the gamma ray seen. The intensity recorded in the table is usually less than that observed in the singles spectrum, because the resolution of the grid chamber is inadequate for complete resolution of the groups and, therefore, the gate contains some contribution from neighboring groups. The major contribution, however, comes from the low energy tailing of the higher intensity groups which as already seen in Figure 49 even for the alpha spectrograph is a problem. The emphasis is, therefore, placed on the variation in intensity with gating position rather than absolute intensities. When no coincident gamma ray was seen this is indicated by a dash. It should be stated that the statistical fluctuation was rather large in these experiments, because the alpha counting rate could not be very high in the alpha grid chamber and, therefore, the coincidence counting rate was low. The counts accumulated in the photopeaks in a one-day run ranged between 10 and 200.

#### d. Decay Scheme

Making use of the data obtained in this research as well as that obtained from other work, the decay scheme (Figure 53a) was constructed in the following way. The observation of the 43 kev gamma ray, and the 56 and 99 kev gammas is in agreement with the transitions previously observed de-exciting the levels at 42.4 and 97.3 kev. The multipolarities of these transitions are tested in Table XIV and indicate the parities of the levels to be the same as that of the ground state.

The  $\alpha_{71}$  group and the 71.4 kev transition was observed by the Russian workers in an intensity of 0.3 per cent (Table XIII and XIV). On the basis of energy and intensity considerations it is most probable that the 71.4 kev transition is associated with the transition from the 71 kev level to the ground state. The multipolarity of this transition was not determined. A consideration of the alpha and transition intensities together with the gamma ray intensity indicates that the transition is either M2 or an E2-M1 admixture. It will be



MU-23342

Fig. 53a. Decay scheme of  $\text{U}^{233}$  (this work).

seen that the latter choice is the more probable (Section e).

Besides noting the gamma rays present when gating on the  $\alpha_{164}$  region (Table XVI), it is of interest to compare their intensities in coincidence with  $\alpha_{164}^+$  and  $\alpha_{164}^-$ . To explain the data, it is necessary to have levels at 118, 146, and 164 kev. The 118 kev gamma ray will be considered first. Its intensity is 0.012 per cent, while its transition intensity from electron spectra is 0.03 per cent with an assignment of E2 (Table XIV). For an E2 transition of this energy, the conversion coefficient is 6.4. Therefore, the intensity of the gamma ray associated with this is about 0.005 per cent. This leaves about half of the intensity unaccounted for, which must be associated with another transition. Now Table XVI shows more of it to be in coincidence with  $\alpha_{164}^-$  than  $\alpha_{164}^+$  and Table XV shows that although it is in full coincidence with the L x-rays only about half of it is in coincidence with the 42 kev gamma ray. This indicates the existence of a level at 118 kev. The fact that this radiation is in full coincidence with L x-rays might be explained by higher lying levels cascading through it. Since this 118 kev transition was not detected in the electron spectrum, the radiation must be E1 and the parity opposite to that of the ground state. Also no other very intense gamma rays or internal transitions could be associated with the de-excitation of this level; therefore, most of the de-excitation must occur by means of the 118 kev transition. This information can then be used to estimate the intensity of the level to be about 0.01 per cent.

There was no transition of 142 kev observed in the electron spectrum although its existence in the gamma spectrum was obvious. The fact that it is only in partial coincidence with L x-rays and the 42 kev gamma radiation and more counts were observed in coincidence with  $\alpha_{166}^-$  than  $\alpha_{166}^+$  indicates a level at 142 kev. This was also suggested in the gamma-alpha coincidence spectrum (Figure 51). The 142 kev transition is probably E1. If it were of a higher multipolarity, it should have been observed in the electron spectrum. This indicates the 142 kev level must be of opposite parity to that of the ground state. Using the same arguments as in the case of the 118 kev level, the alpha intensity to the state is estimated to be 0.01 per cent.

Table XIV indicates that no 162 kev radiation was detected in the electron spectrum, although a gamma ray of this energy was observed. From Table XVI it is seen to be more in coincidence with  $\alpha_{166}^{+}$  than  $\alpha_{166}^{-}$ ; Table XV shows it to be in partial coincidence with L x-rays and the 42 kev gamma ray. The gamma-alpha coincidence shows a very pronounced peak at the appropriate energy. This is interpreted as a 162 kev transition de-exciting a level at 162 kev to the ground state. Its absence in the electron spectrum strongly suggests E1 radiation. Proceeding as before, the intensity of the 162 kev level is estimated to be about 0.01 per cent. From Table XIII, the intensity of  $\alpha_{164}$  is seen to be 0.05 per cent. In the internal conversion electron spectrum (Table XIV) there are observed transitions of 66.0 and 121.0 (E2) which have been indicated as de-exciting to the 164 kev level to the 97.3 and 42.4 kev levels respectively.<sup>69</sup> As already stated there was observed 118 kev gamma radiation which in part is associated with this 121.0 kev transition. The fact that the 163.4 kev level de-excites by means of E2 radiation to a level which in turn de-excites to the ground state by M1 - E2 radiation, while the 162 kev gamma ray is most probably E1 indicates the existence of two levels of opposite parity. Since the intensity of the 162 kev level is estimated to be 0.01 per cent, the 163.4 kev level is about 0.04 per cent.

A gamma ray of 73 kev is seen to be more in coincidence with  $\alpha_{166}^{-}$  than  $\alpha_{166}^{+}$ , and also in full coincidence with L x-rays and 42 kev gamma ray while it is in partial coincidence with the 57 and 95 kev gamma rays (Tables XV and XVI). As previously stated a transition of about the same energy in an intensity of 0.3 per cent was detected in the electron spectrum. It is obvious then that only part of the 73 kev gamma radiation is associated with the transition seen in the electron spectrum. The rest of the 72 kev gamma radiation must de-excite either the level at 118 kev or 142 kev to the 42 and 71 kev levels respectively or both. The fact that the 72 kev gamma radiation is in full coincidence with L x-rays and 42 kev gamma ray would indicate higher lying levels cascading through this one. The multipolarity of this radiation must be E1 on the basis of intensity coincidences. This is in agreement with the parity assignments of the levels at 118 and 142 kev.

Radiation of 211 kev was not observed in the electron spectrum. A gamma ray of this energy and 0.006 per cent intensity was observed in the gamma spectrum. A 211 kev gamma ray was in coincidence with  $\alpha_{211}$  and in the gamma-alpha coincidence spectrum (Figure 51) there was a substantial peak at this energy. From Table XV it is seen that the 211 kev gamma ray is in partial coincidence with L x-rays and nothing else. This is evidence for a state at 211 kev. This gamma ray must be electric radiation because of the low intensity of K x-rays associated with the transition. Since the transition was not seen in the electron spectrum, it must be E1 or E2. In the alpha spectrum obtained by Dzhelepov et al. (Table XIII) a limit was set on  $\alpha_{234}$  as  $< .007$  per cent. If  $\alpha_{211}$  were greater than this, it should have been seen. Therefore, if  $\alpha_{211}$  were E2, the alpha population to that level would be 0.011 per cent, clearly exceeding the alpha intensity limit. It must therefore be E1, which indicates a parity for this state opposite to that of the ground state.

In coincidence with  $\alpha_{211}$  there were also seen some gamma rays of 72, 117, 142 (really about 146 kev in the coincidence spectra) and 162 kev. From this it is apparent that some de-excitation takes place to the levels at 162, 142, and 118 kev, either directly or through cascade. The intensity of the 211 kev level is estimated to be about 0.01 per cent.

In the gamma-alpha coincidence spectrum the complex group at  $\alpha_{220}$  was graphically resolved into  $\alpha_{211}$  and  $\alpha_{245}$ . In Table XIII the spectrum of Dzhelepov indicated there might be a level populated by  $\alpha_{240}$  in an intensity less than 0.007 per cent. In the coincidence spectra no gamma rays were detected which could be attributed to the de-excitation of a level of this energy. It must de-excite by a series of cascade transitions.

Interest will now be directed to the 300 kev region. In the alpha spectrum (Table XIII) there is abundant evidence for a state at 317 kev; its intensity is 0.04 per cent. In the gamma-alpha coincidence spectrum this group dominated the lower portion of the spectrum (Figure 51). A gamma ray of this energy in an intensity of 0.01 per cent is clearly in evidence (Table XIV). In Table XV it is seen to be in coincidence with no other gamma radiation, and in Table XVI it is in

coincidence with  $\alpha_{320}$ . This gamma ray is definitely associated with the decay of the 317 keV level to the ground state.

Besides the 318 keV radiation, 242 and 282 keV gamma rays were seen in the gamma spectrum as well as the  $\alpha_{320}$ -gamma coincidence spectrum. From the electron spectra (Table XIV) the following transitions were observed: 245.3 keV (0.015%), 248.6 keV (0.008%), 277.8 keV (0.005%), 291.5 keV (0.012%), 317.0 keV (0.02%), and 321.0 keV (0.008%). The multipolarities could not be determined from experiment for the 245.3, 291.5 and 317.3 keV transitions, but were considered to be M1 on the basis of intensity and plausibility arguments.<sup>69</sup> It will be shown that these assignments are consistent with the data obtained in this research.

The possible multipolarities for the 245.3 keV transition are M1, M2, E1 and E2, for which cases gamma ray intensities of 0.011 per cent, 0.003 per cent, 0.05 per cent and 0.25 per cent are expected. For the 248.6 keV transition, the same multipolarities with the intensities of the associated gamma rays being 0.005 per cent, 0.0013 per cent, 0.027 per cent and 0.13 per cent for M1, M2, E1 and E2, respectively. The experimental data on the 242 keV gamma ray indicate that it is associated with the decay of the 318 keV level, and thus it is assumed to be associated with the two transitions of about this energy observed in the electron spectrum. Its intensity of 0.005 per cent definitely rules out electric multipole transitions. The best agreement would be obtained if the 248.6 keV transition is M1 and the 245.3 keV transition is M2. However, it will be shown shortly that the electron transition intensities are too high by a factor of 3 and, therefore, both transitions are in agreement with M1 assignments.

The same arguments can be advanced for the 277.8 and 291.5 keV transitions as well as the 317.0 and 321.0 keV transitions. The conclusions are also the same, namely that the 277.8 and 321.0 keV transitions are M1 and the 291.5 and 317.0 keV transitions M2. These transitions come from either two levels at 321.0 and 317.0 keV of total intensity  $0.033 \pm 0.008\%$ ; or from three levels at 291.5, 317.0 and 366.0 keV of total intensity 0.05 per cent (alternate decay scheme to be discussed in Section 5). The gamma ray intensities total 0.022 per cent while the electron intensities total 0.68 per cent for the former and 0.023 per cent and 0.073 per cent respectively for the latter.

In either case it is seen that the electron intensities are approximately a factor of 3 too high. Therefore, reconsideration of the previous arguments shows that M1 assignments for the six transitions are in agreement with the data.

Having established the multipolarities of these transitions, it is now necessary to fit these transitions into the decay scheme. It is obvious that the six transitions considered above cannot all de-excite the 317 kev level. The gamma-alpha coincidence spectrum (Figure 51) shows definite indications of two more alpha groups: one at 367 kev and one at 288 kev. As seen in Table XIII and XIV, there is definite evidence for a level at 367 kev from alpha, electron, gamma, and coincidence spectra. The data obtained in this research together with the previous data confirm the existence of this level. It is fairly certain that the 370 kev gamma ray and 366 kev transition observed in the electron spectrum belong to the same transition and de-excite this level to the ground state. The level at 288 kev is tentative but will be shown to fit into the decay scheme thus lending support to its existence.

The gamma-gamma coincidence data (Table XV) indicated that the 282 kev gamma ray is in partial coincidence with L x-rays and nothing else. Therefore, either the 277.5 kev or the 291.5 kev transition must go directly to the ground state. In view of the small energy difference between the proposed level at 288 kev and 291.5 kev, this transition is placed as de-exciting the level (taken as 291 kev) to the ground state. The 248.6 kev transition agrees well with the energy difference between the levels 291.5 kev and 42.4 kev. Placement of the transition in this position is in agreement with the coincidence data.

The 277.5 kev transition is within the experimental energy difference between the 317.0 and 42.4 kev levels. Placement in this position would also explain the partial coincidence of the 282 kev gamma with L x-rays (Table XVI).

The energy of the 245.3 kev transition agrees well with the energy difference between the 317.0 and 71.4 kev levels. This is in agreement with the L x-ray-gamma coincidence i.e. both the 245.3 and 248.6 kev transitions which are included in the 242 kev gamma ray were in full coincidence with L x-rays. It does not explain the full coincidence between the 42 and 242 kev gamma rays. In the proposed

decay scheme there should only be a partial coincidence. This difficulty could be resolved by proposing the existence of a new level at 287.7 kev. However, in doing this the intensity of the group seen in the gamma-alpha coincidence spectrum would be nearly a factor of 2 greater. One reason for the full coincidence then might be that there is enough contribution from the escape peak of the 72 kev gamma ray to cause this effect so that actually some of the coincidence may be due to a 72 kev gamma coincidence.

The 321.0 kev transition agrees with the energy difference between the 365.0 and 42.4 kev levels within experimental error. It is placed in this position in the decay scheme. The seven highest energy transitions just considered are most probably M1. This requires that the levels 71.4, 291.5, 317.0, and 366.0 have the same parity as the ground state.

The alpha decay data are summarized in Table XVIII. The states not enclosed by parenthesis indicate states observed to be populated directly in the alpha spectrum, a single set of parenthesis indicates those levels observed in all types of coincidence spectra; a double set indicates those groups observed only in alpha-gamma and gamma-gamma coincidence.

The proposed decay scheme is shown in Figure 53a. The levels considered certain are shown by a solid line; those considered less certain are shown by a dashed line. For comparison, the decay scheme of Tret'yakov is also shown (Figure 53b).

Besides the odd parity states, differences between the two decay schemes occur at the levels 29.1, 320.0 and possibly 131 kev, and the 291.5 kev level in this research. On the basis of this research, nothing can be said about the existence of the 29.1 kev and the possible 131 kev levels. They would not have been observed in this work, nor were they needed to explain the data. The 320 kev level was proposed by them to explain the existence of 4 transitions. However, it was seen that these transitions would be explained just as well if a level at 291 kev is proposed together with the 317 and 365 kev levels. In addition, the coincidence data strongly suggest the presence of a level at this energy.

A check of the intensity balance of the two decay schemes would be informative if the limits of error of the alpha and internal

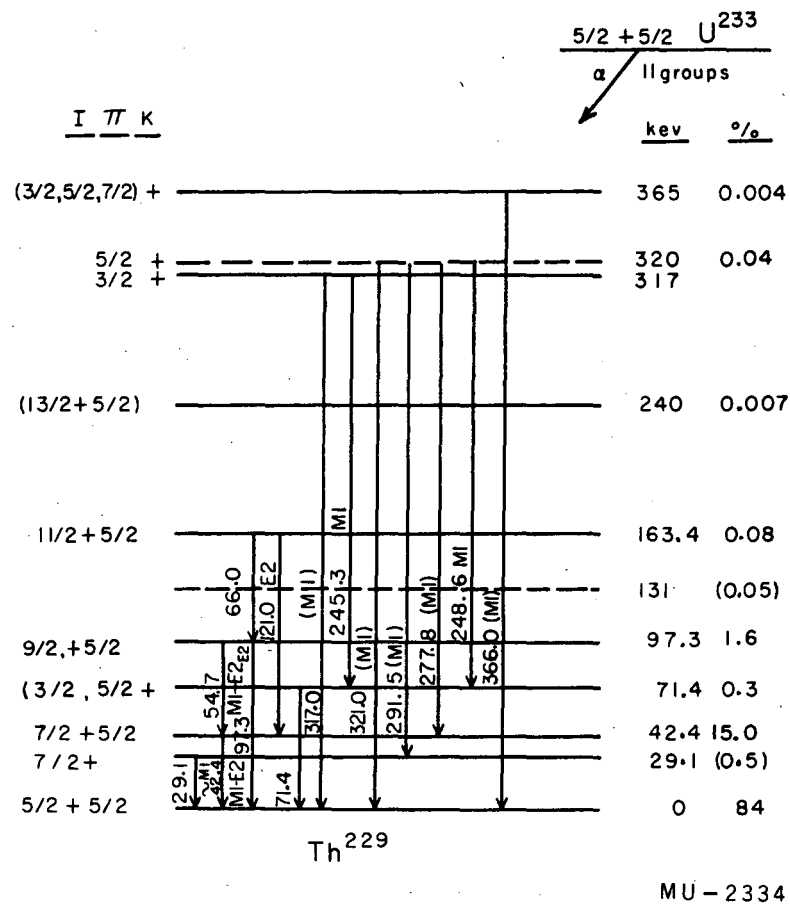


Fig. 53b. Decay scheme of  $\text{U}^{233}$  (Tret'yakov et al.)

transitions intensities were smaller. With the present data such a comparison would not be a very good test.

The only gamma ray unaccounted for is the 182 kev gamma ray whose existence is not certain. It was not placed in the decay scheme since there is no obvious place for it.

TABLE XVII.

| Alpha Groups of $U^{233}$     |                          |                  |
|-------------------------------|--------------------------|------------------|
| Excited State<br>Energy (kev) | Abundance % <sup>a</sup> | Hindrance Factor |
| 0                             | 83.7                     | 1.17             |
| 43 ± 1                        | 14.1 ± 0.7               | 3.66             |
| 99 ± 2                        | 1.9 ± 0.3                | 1.05 x $10^1$    |
| ((118)) ± 5                   | 0.01                     | 1 x $10^3$       |
| (142) ± 5                     | 0.01                     | 1 x $10^3$       |
| (162) ± 5                     | 0.01                     | 7 x $10^2$       |
| 163 ± 3                       | 0.04 ± 0.03              | 2.0 x $10^2$     |
| (211) ± 3                     | 0.01                     | 4 x $10^2$       |
| (245)                         | 0.01                     | 2 x $10^2$       |
| (291) ± 3                     | 0.01                     | 9 x $10^1$       |
| 317 ± 2                       | 0.04 ± 0.02              | 1 x $10^1$       |
| (365) ± 4                     | 0.004 ± 0.003            | 6 x $10^1$       |

<sup>a</sup> The intensities of those groups enclosed by parenthesis, with the exception of  $\alpha_{365}$ , should be regarded approximate and represent upper limits in some cases.

#### e. Interpretation

##### (1) Even Parity States:

The spin of  $U^{233}$  has been measured to be  $5/2$ ,<sup>70</sup> and has been shown to be the state  $5/2 + (6,3,3)$ . The levels at 42.4, 97.3, and 163.4 have already been interpreted as rotational members based on the ground state assignment of  $I = K = 5/2$ .<sup>69</sup> Using Equations 3 and 4 the energies and intensities of the members of this band were calculated and are tabulated in Table XVIII. The experimental energies were from the electron work and the intensities are an average of those obtained in this work and those of Dzhelepov et al. The intensity of  $\alpha_{163}$  was obtained by subtracting off the contribution from the odd parity state of almost this same energy. These data, as well as the mode and multipolarities of the transitions de-exciting these levels confirm such an interpretation.

It has already been shown that the 291, 317 and 365 kev states have the same parity as the ground state (Section d). Since these levels decay to states of spin  $5/2+$ , and  $7/2+$  by M1 radiation, their spins and parities must also be  $5/2+$  or  $7/2+$ .

For the 317 kev level a spin of  $5/2$  is preferred. This spin would allow the state to be populated by  $l = 0, 2, 4$  alpha waves which could account for the relatively low hinderance factor. This assignment is also preferred on the basis of the ratios of the reduced transition probabilities to the two lowest states of the  $K = 5/2$  band.

Assuming the 317 kev state to be  $I = K = 7/2$ , then the ratio

$$\frac{B(M1; 7/2 \rightarrow 5/2)}{B(M1; 7/2 \rightarrow 7/2)} = 3.4; \text{ for the case } I = K = 7/2, \text{ then}$$

$$\frac{B(M1; 5/2 \rightarrow 5/2)}{B(M1; 5/2 \rightarrow 7/2)} = 2.4.$$

The experimental value is 2.5, clearly indicating the latter assignment.

This suggests that the 365 kev state may be a rotational satellite of this level in which case its spin would be  $7/2$ . The calculated reduced M1 transition probability of cascade to crossover is 1.9. The experimental value is 2.6. The 366 kev electron transition intensity from this level would have been difficult to determine because only the K line of this transition was observed as a broadening on the high energy side of the K line of the 317 kev transition. It, therefore, could have been slightly overestimated. If its intensity is decreased from 0.008 per cent to 0.007 per cent then the ratio becomes 2.0 in much better agreement.\*

This is to be compared with the agreement obtained from the alternate decay scheme. The theoretical ratio assuming  $I = K = 5/2$  for cascade to crossover transition from the 321.0 kev level is 0.40; for  $I = 5/2, K = 3/2$  the ratio is 2.5. The experimental value is 1.0.

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\* A transition to the  $9/2+$  state should occur if the 365 kev state is really  $7/2$ . Its reduced transition probability is nearly the same as the transition going to the  $7/2$  state. However, its intensity would be a factor of 2 less because of the smaller energy. This could account for its apparent absence in the spectrum.

TABLE XVIII

Energies and Intensities to  $K = 5/2$  Band

| Spin of State | Excited-State Energy |        | Relative Intensities |         |
|---------------|----------------------|--------|----------------------|---------|
|               | Theor. <sup>a</sup>  | Exper. | Theor. <sup>b</sup>  | Exper.  |
| 5/2           | (0)                  | 0      | (100)                | 100     |
| 7/2           | (42.4)               | 42.4   | 13                   | 17.0    |
| 9/2           | 96.9                 | 97.3   | 1.8                  | 2.1     |
| 11/2          | 163.6                | 163.4  | 0.1                  | 0.05    |
| 13/2          | 242.3                |        | 0.01                 | < 0.008 |

<sup>a</sup> Experimental values used to evaluate rotational constant  
 $\hbar^2/2\mathcal{I} = 6.057$  kev.

<sup>b</sup>  $c_0 = 1.00$ ,  $c_2 = 0.87$ ,  $c_4 = 0.07$

That the reduced transition probabilities are described accurately in this region by theory is seen in the electron capture decay of  $\text{Pa}^{233}$  to  $\text{U}^{233}$ .<sup>71</sup> For the transition from the  $K = 3/2$  to  $K = 5/2$  band the theoretical value for  $\frac{B(M1; 5/2 \rightarrow 7/2)}{B(M1; 5/2 \rightarrow 5/2)}$  is 2.5 while the experimental values are 2.3 and 2.1.

The only Nilsson state of  $I = K = 5/2$  in this vicinity is the state  $5/2 + (6,2,2)$  (Figure 14). From the diagram, it is seen that at a deformation of  $\delta = 0.22$  the  $1/2 + (631)$  state is slightly below it in energy. At smaller deformations the two states cross with the  $1/2 +$  state rising above the  $5/2 +$  state. The  $1/2 +$  level has been observed at 400 kev in  $\text{U}^{233}$ . Since the deformation is smaller in the  $\text{Th}^{229}$  then it is not inconceivable that the  $5/2 + (622)$  level would be at 317 kev. This assignment is tentatively made for this level. If the 365 kev level is the first excited state based on the 317 kev level then the rotational constant would be 6.9 kev. In  $\text{U}^{233}$  the rotational constants are 5.76 kev ( $K = 5/2$  band), and 7.41 kev ( $K = 1/2$  band).

The level at 291 kev must have a spin and parity of  $5/2 +$  or  $7/2 +$  since it decays to levels of these spin and parities by M1 radiation. A glance at Figure 14 shows that there is no Nilsson state to which this can reasonably be assigned.

As seen in Section d, the 71.4 kev level is of positive parity. Since M1 radiation goes to this state from the 317 kev

$5/2+$  state, its spin can be  $3/2 \leq I \leq 7/2$ . From the Nilsson diagram (Figure 1) the only reasonable assignment for this state is  $3/2+$  (631). This state has been observed in  $U^{233}$  at 312 kev (141 neutrons) and very probably as the ground state in  $U^{229}$  (137 neutrons) (Section A). For a nucleus of 139 neutrons a  $3/2+$  level is therefore expected between 0 and 313 kev; the 71.4 kev level appears to be this level.

## (2) Odd Parity States:

As already stated (Section 1), the four levels at 118, 142, 162, and 211 kev are of negative parity. From the Nilsson diagram it is seen that there are two negative parity states in this region  $5/2-$  (752) and  $7/2-$  (744). In  $Th^{231}$ , the  $5/2-$  state lies below the  $7/2-$  (185 and 390 kev respectively). In  $U^{233}$  the  $5/2-$  is thought to be at 300 kev.<sup>31</sup> It, therefore, is reasonable that the negative parity states observed in  $Th^{229}$  are rotational members of these intrinsic states. The close proximity of these bands where  $K$  differs by one unit would be expected to produce a considerable perturbation of the band members. It should be noted that this is not inconsistent with the data obtained from the electron capture decay of  $Pa^{229}$  (Section 2) although from these data more specific assignments cannot be made.

## 2. Electron Capture Decay of Protactinium-229

$Pa^{229}$  was first investigated by Hyde and Studier who found it to decay with a 1.5 day half-life.<sup>72</sup> Meinke et al.<sup>73</sup> confirmed this half-life and determined the electron capture branching ratio to be about 99 per cent. The later data of Slater and Seaborg indicated the electron capture branching ratio to be 0.25 per cent.<sup>74</sup> Since that time a study of the conversion electrons accompanying the electron capture decay by Hok<sup>75</sup> and by Hill<sup>8</sup> indicated the presence of one transition of 42 kev. From the relative subshell ratios Hill determined the transition to be 95% E2 - 5% M1, but no estimate could be made of the population to the state giving rise to the 42 kev transition. No other transitions were observed which would be ascribed to  $Pa^{229}$ , neither in the conversion electron study nor in the gamma ray studies. The difficulty associated with the above work was that  $Pa^{229}$  had to be studied in the presence of other protactinium isotopes produced in the bombardment. Any weak transitions occurring in the  $Pa^{229}$  decay could then easily be obscured by them.

The present study of  $\text{Pa}^{229}$  was initiated for two reasons:

(a) a small amount of activity could be readily obtained isotopically pure from the electron capture decay of  $\text{U}^{229}$ ; (b) since the decay energy is about 280 kev (estimated from closed cycle decays), then there existed the possibility of levels other than the 42 kev level populated in the decay. The observation of transitions from these levels would therefore aid in the understanding of the complex level structure existing in  $\text{Th}^{229}$  as revealed from the alpha decay of  $\text{U}^{233}$  (Section 1).

a. Gamma Spectra

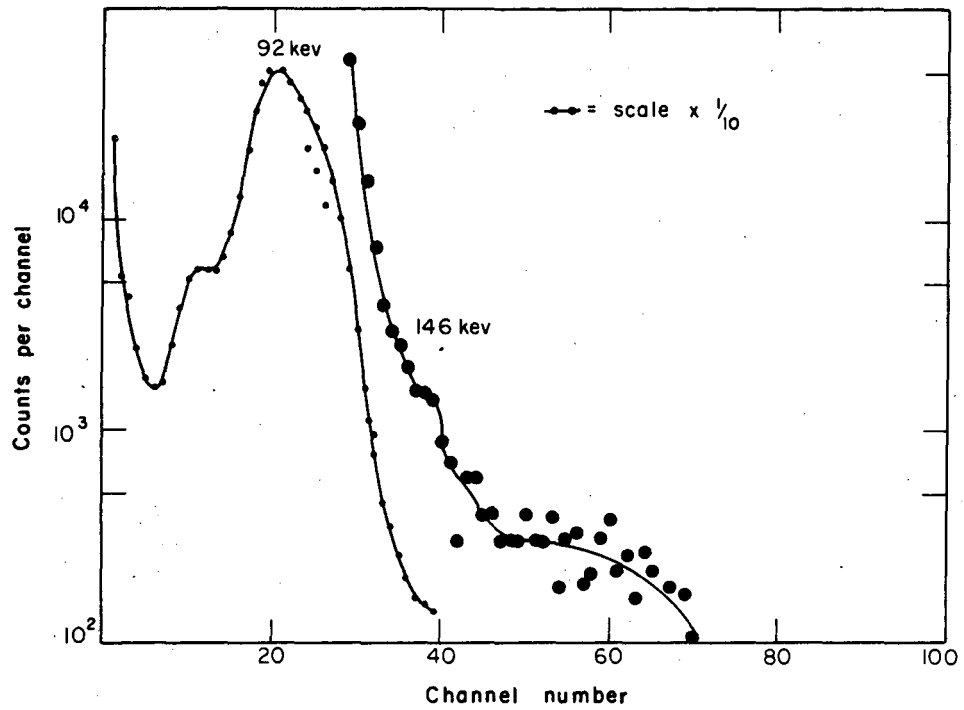
The singles spectra showed the presence of L and K X-rays together with a low intensity shoulder at 146 kev and a lower intensity continuum extending to higher energies (Figure 54). This continuum was attributed to radioactive impurities in the source. The decay of the X-rays was followed and found to exhibit a 1.5 day half-life.

L X-ray - gamma and K X-ray - gamma coincidence experiments were performed (Figures 55-56). In the L X-ray gamma coincidence experiments, the spectra showed K X-rays in an intensity of 90 per cent and a 147 kev gamma ray of 0.14 per cent intensity. Higher energy radiation was observed but its intensity changed relative to the K X-ray intensity. Most of it was thus not due to  $\text{Pa}^{229}$  decay and an upper limit of 0.02 per cent could be set for the intensity of transitions of energy greater than 150 kev.

The K X-ray - gamma coincidence spectra showed radiation in the K x-ray region in an intensity of 2 per cent and also the 148 kev gamma ray in an intensity of 0.10 per cent. Again the higher energy continuum radiation was present which was probably all due to contaminants.

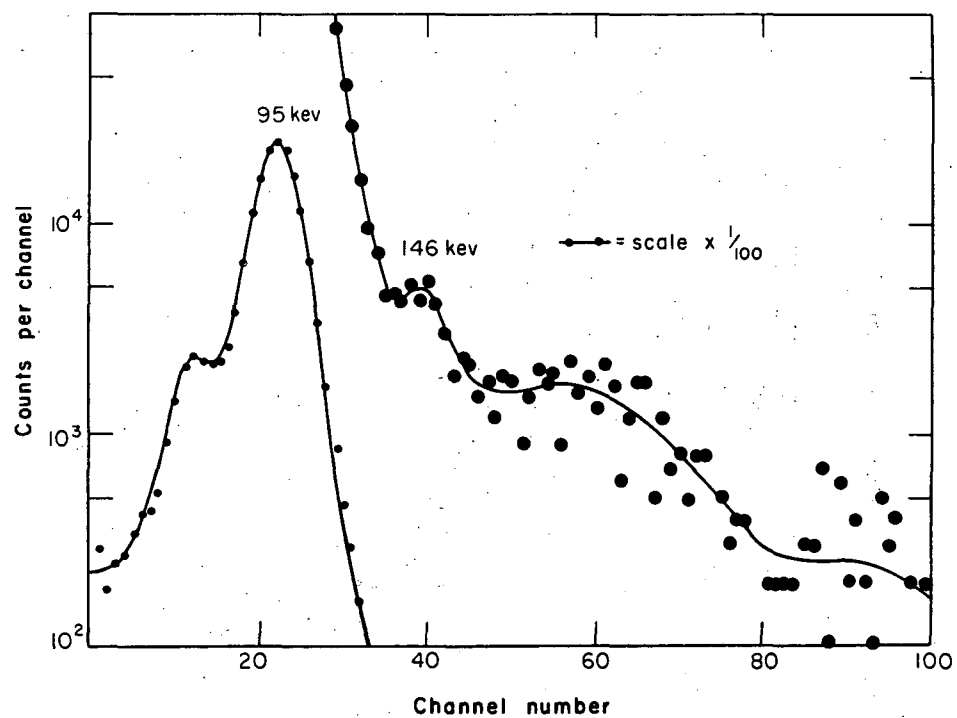
b. Interpretation

Since the energy available for electron capture in  $\text{Pa}^{229}$  was calculated to be 0.28 Mev, states in  $\text{Th}^{229}$  populated by K-electron capture cannot be higher in energy than 0.17 Mev and those populated by L-electron capture must be less than 0.26 Mev. Since the 147 kev gamma ray was in coincidence with both L and K x-rays it must de-excite a level lying in the energy region  $147 \leq E \leq 170$  kev. As observed previously in the decay of  $\text{U}^{233}$  (Section 1) a level has been observed at 142 kev and was found to emit a gamma ray of this energy. It is



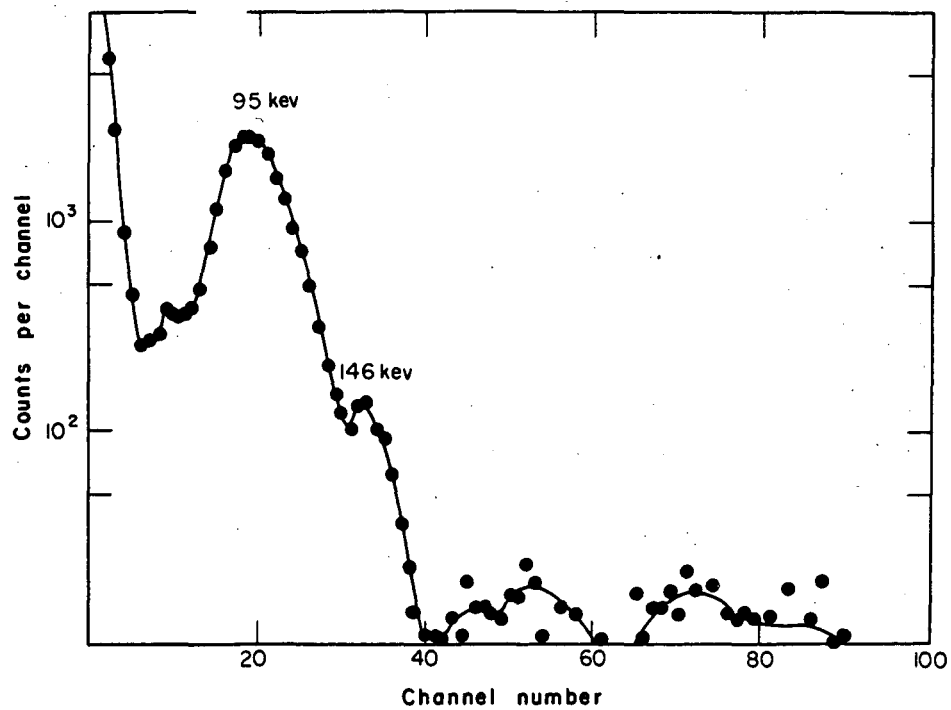
MU-23344

Fig. 54. Gamma ray spectrum of  $\text{Pa}^{229}$ .



MU-23345

Fig. 55. L X-ray—gamma ray coincidence spectrum of  $\text{Pa}^{229}$ .



MU-23346

Fig. 56. K X-ray-gamma ray coincidence spectrum of  $\text{Pa}^{229}$ .

therefore highly probable that the 146 kev gamma radiation observed in the Pa<sup>229</sup> decay de-excites this same level. Arguments concerning the nature of this state have been given in Section 1-e.

It is not possible to account for the radiation observed at 95 kev in coincidence with K x-rays in the Th<sup>229</sup> level scheme. The fact that its intensity fluctuated on experiments with different sources suggests that most of it was due to the presence of impurities (the counting rate was not high enough to account for this by chance coincidence events).

As stated previously, Hill determined the 42 kev transition to be 95 per cent E2 - 5 per cent M1. Since this presumably arises from de-excitation of the 42 kev level, then the mixing ratio should be the same as determined from U<sup>233</sup> decay. In this case, however, the ratio was 20 per cent E2 - 80 per cent M1 (Table XIV). Since the mixing for the analogous transition in U<sup>233</sup> (40 kev) is 30 per cent E2 - 70 per cent M1, it would appear that the correct mixing ratio is 20 per cent E2 - 80 per cent M1. Although these experiments were designed specifically to observe gamma radiation in the decay of Pa<sup>229</sup>, a very rough number was calculated for the population to the 43 kev level from these data. First of all, it was noted that no 43 kev gamma ray was observed in the K x-ray - gamma coincidence spectrum. The lower limit of detection was about 0.2 per cent which placed an upper limit of 60 per cent for the population to the state. It has already been mentioned that the decay of the L and K x-rays was followed, with the L x-rays being counted with one detector and the K x-rays with the other. Therefore, L and K x-rays did not appear in the same spectrum which introduced another variable into the following calculation. From the ratio of L to K-shell vacancies<sup>76,77</sup> was subtracted the number of L-shell vacancies produced by a K-shell vacancy, 0.72.<sup>78</sup> The resulting number then included the ratio of L-electron capture and L conversion from the 42 kev transition to K-electron capture. The L to K-electron capture ratio was estimated to be about 0.4 assuming a first forbidden transition.<sup>79</sup> After subtraction, the resulting quantity gave about 20 per cent for the population to the 43 kev level.

The population to the 42 kev state was also calculated from the L x-ray gamma coincidence spectrum. The number of K x-rays in coincidence with L x-rays was found to be 0.90. Contributions to this

number come from coincidence between the L vacancies produced by the K vacancies and the L vacancies produced by L-electron capture and from the conversion of the 43 kev transition populated by L-capture. After subtraction of the various contributions, the population to the state was calculated to be 50 per cent.

These estimates give an order of magnitude number of 30 per cent. This intensity does not conflict with the value calculated for first forbidden, unhindered transition with  $\Delta I = 1$ . The 30 per cent population corresponds to a log ft value of 6.

### 3. Radium-222 Alpha Spectrum

The alpha spectrum of  $\text{Ra}^{222}$ , studied in equilibrium with its grandparent  $\text{U}^{230}$ , was obtained on an alpha particle spectrograph. Under these conditions only the  $\text{Ra}^{222} \alpha_0$  group was observed; the  $\alpha_{325}$  group, indicated by the presence of a 325 kev gamma ray, was obscured by the  $\text{Th}^{226} \alpha_{112}$  alpha group which occurs at about the same energy. The intensity of the  $\text{Ra}^{222} \alpha_{325}$  group was estimated from the intensity of the gamma ray for an E2 transition to be  $4.4 \pm 1$  per cent.<sup>54</sup>

During the course of the experiments with the  $\text{U}^{229}$  family,  $\text{U}^{230}$  was made as a by-product. The same technique was employed to obtain an alpha spectrum of  $\text{Ra}^{222}$  as was used for  $\text{Ra}^{221}$ . The spectrum, obtained with the alpha particle grid chamber, is shown in Figure 57. A group, occurring at an energy of  $321 \pm 4$  kev below the  $\text{Ra}^{222} \alpha_0$  peak, is clearly defined in good agreement with the gamma ray energy. (The scale was calibrated using the  $\text{Th}^{226} \alpha_0$  and  $\text{Ra}^{222} \alpha_0$  groups from a  $\text{Th}^{226}$  sample.) The intensity of the group is  $3.2 \pm 0.4$  per cent, about 1 per cent lower than the previously reported value. This then makes the population to the ground state 96.8 per cent. This also increases the intensity of the  $\text{Th}^{226} \alpha_{112}$  state 1 per cent which decreases the intensity of the  $\text{Th}^{226} \alpha_0$  state.

Making use of the fact that the 325 kev transition is E2, the intensity of the gamma ray was calculated to be 2.7 per cent. This agreed with the experimentally observed intensity of  $2.9 \pm 0.5$  per cent determined in conjunction with the alpha-gamma coincidence experiments on the  $\text{U}^{229}$  family. This is 22 per cent lower than the 3.6 per cent previously reported (but within their lower limit of error). Since the intensities of all the other gamma rays of the  $\text{U}^{230}$

family were determined relative to this one, then they also are too high and thus have been reduced by this same factor. These results are presented in Tables XX and XXI, together with the previously reported values.

TABLE XIX

| Alpha Group                        | Intensity %   |           |
|------------------------------------|---------------|-----------|
|                                    | Previous Work | This Work |
| Th <sup>226</sup> α <sub>0</sub>   | 79            | 78        |
| Th <sup>226</sup> α <sub>112</sub> | 19 ± 1.5      | 20        |
| Ra <sup>222</sup> α <sub>0</sub>   | 95            | 96.8      |
| Ra <sup>222</sup> α <sub>325</sub> | 4.4 ± 1       | 3.2 ± 0.4 |

TABLE XX

| Isotope           | Gamma Ray Energy (kev) | Intensity %   |                        |
|-------------------|------------------------|---------------|------------------------|
|                   |                        | Previous Work | This Work <sup>a</sup> |
| U <sup>230</sup>  | 72                     | 0.75 ± 0.11   | 0.59                   |
|                   | 158                    | 0.33 ± 0.06   | 0.26                   |
|                   | 232                    | 0.24 ± 0.05   | 0.19                   |
| Th <sup>226</sup> | 112                    | 4.8 ± 0.4     | 3.8                    |
|                   | 131                    | 0.4 ± 0.1     | 0.3                    |
|                   | 197                    | 0.4 ± 0.05    | 0.3                    |
|                   | 242                    | 1.2 ± 0.1     | 0.9                    |
| Ra <sup>222</sup> | 325                    | 3.6 ± 0.7     | 2.9 ± 0.5              |
| Em <sup>218</sup> | 609                    | 0.2 ± 0.05    | 0.15                   |

<sup>a</sup> No limits of error are placed on these values (with the exception of the 325 kev gamma ray) because the intensities were not experimentally determined in this work. Therefore the limits of error are those of the previous work.

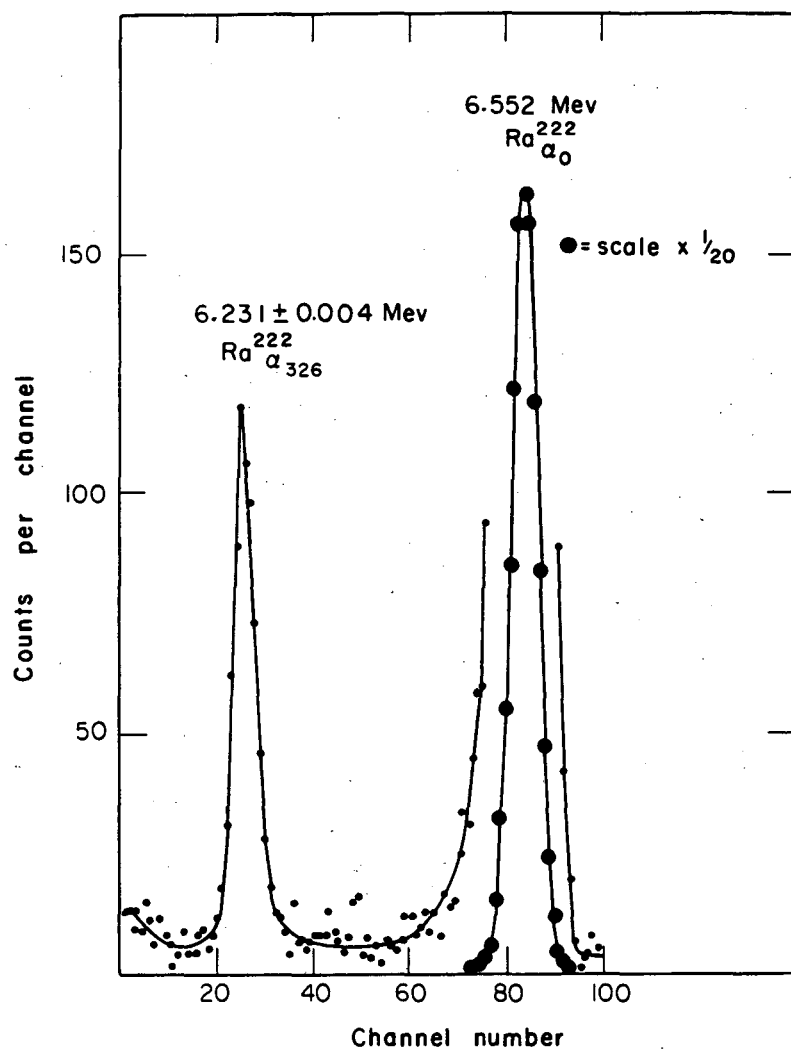
#### 4. Radon-218 Half-Life Determination

The previously reported value for the half-life of Rn<sup>218</sup> was 0.019 seconds. This was measured by visually observing the time interval between two successive pulses on an oscilloscope with a sweep time of 0.2 seconds. The pulses were produced by the alpha disintegration from a U<sup>230</sup> source in equilibrium with its daughters.<sup>80</sup>

The present determination made use of the apparatus described

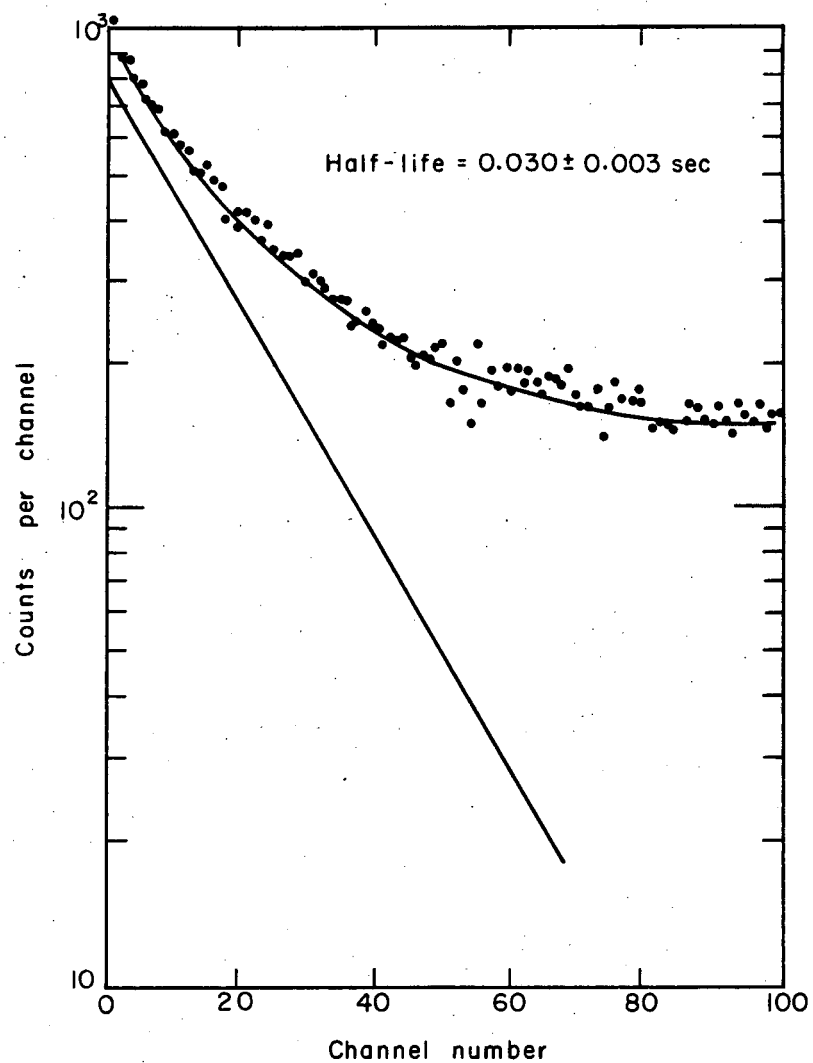
in Section II-C-1. A  $U^{230}$  source was used to supply the  $Ra^{222}$  and  $Rn^{218}$ . One single-channel analyzer was set to accept  $Ra^{222}$  pulses to trigger the circuit, while the signal pulses were obtained from the single-channel analyzer set to pass the  $Rn^{218}$  alpha pulses. Oscilloscope sweep times of 0.2 and 0.5 seconds were used. Because of the relatively long sweep time and the fact that the source geometry was only 50 per cent, count rates of 40 counts per minute had to be used to keep the true-to-chance coincidence ratio reasonably high. Several determinations were made each of which took about 2 days. A typical decay curve, shown in Figure 58 was obtained using a 0.2 second sweep time. In all cases, chance coincidence spectra were obtained following each run. These spectra were then subtracted from the decay curves after being normalized at the tail. In this way any deviations from linearity could be corrected for.

The half-life obtained for the decay of  $Rn^{218}$  was  $0.030 \pm 0.003$  seconds. This is in better agreement with the half-life-energy relation.<sup>41</sup>



MU-23347

Fig. 57. Alpha spectrum of  $\text{Ra}^{222}$  taken with alpha particle grid chamber.



MU-23349

Fig. 58. Delay coincidence curve of  $\text{Rn}^{218}$ .

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