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ALPHA-DECAY STUDIES IN THE HEAVY-ELEMENT REGION

John Philip Hummel

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John Philip Hummel

Radiation Laboratory and Department of Chemistry
University of California, Berkeley, California

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ABSTRACT

Using primarily a 75-cm radius of curvature 60° symmetrical electromagnetic analyzer, a study of the complexity of the following alpha spectra was made: E^{253}, Cf^{246}, Cm^{244}, Am^{243}, $Pu^{236,242}$, Pa^{231}, $Th^{227,230}$, Ac^{225}, At^{209}, and Po^{206}. An investigation of the gamma rays associated with the following isotopes was also made: Cf^{246}, $Cm^{242,244}$, Am^{243}, Pu^{236}, and Pa^{231}.

Decay schemes have been suggested for most of the isotopes included in this study. Those for the even-even isotopes were found to conform well with the previously existing systematics for that group of nuclides. Many of the decay features of the odd-mass isotopes seemed to conform well with presently expanding theories.

I. INTRODUCTION

In the past few years there has been much experimental and theoretical interest in the properties of nuclear energy levels. The hope is that a complete understanding of nuclear energy levels will culminate in an understanding of the nucleus itself. Although advances towards understanding of the various nuclear states have been made, one cannot say that anything near complete understanding has been achieved.

One of the most fruitful regions of study is the heavy-element region (elements beyond lead in the periodic table). A decay property almost exclusive to this region is that of alpha-particle emission, and much of the study of the energy levels in this region has been through analysis of alpha-decay schemes. Regularities in the low-lying energy levels observed in the alpha decay of even-even isotopes have been well established.[1-5] Odd-mass isotopes, however, have not shown the same degree of regularity, although some features seem to be common to them. Also, regularities in the breakdown of simple alpha-decay theory applied to certain transitions in the even-even nuclei have been established.[6] Again, the odd-mass isotopes have not shown such regular deviations.

This research has been conducted to extend the observations noted above for the even-even isotopes and to obtain additional data for the odd-mass nuclei in the hope that more regularities will become apparent for that class of nuclides.

II. EXPERIMENTAL TECHNIQUES

The study of nuclear decay has been called nuclear spectroscopy because of the somewhat analogous approach to the atomic systems. On the basis of the various spectra observed, conclusions are drawn regarding the nature of the various levels involved. The spectra referred to are those of the characteristic nuclear radiations--beta and gamma rays, and alpha particles. The majority of the work done in this particular investigation was concerned with alpha-particle and gamma-ray spectroscopy.

A. Alpha-Particle Spectrographs

Two alpha-particle spectrographs were used in this study. The majority of the alpha spectroscopy was performed on the so-called "low geometry" spectrograph. In recent months, certain alpha-gamma coincidence studies were made with the aid of a new double-focusing spectrograph. The two instruments are discussed separately.

The "low geometry" alpha-particle spectrograph has been described in the literature,[7] but the most important features are presented here. It is a converted Nier type spectrograph employing a 60^{o} sector magnet. The radius of curvature of the normal trajectory is 75 cm. The magnetic field over the 1-inch gap between the pole pieces is produced by an electromagnet capable of bending a 14-Mev alpha particle at maximum field intensity. Figure 1 is a schematic drawing of the spectrograph showing the relative positions of the source, magnet, and receiver units. The entire unit is evacuated to an operating vacuum of about 10^{-5} mm of mercury prior to operation.

One of the most important considerations of an electromagnet spectrograph is the stability of the magnet current supply. The magnet power supply on this spectrograph is capable of maintaining the current constant to one part in 10,000 over at least a 24-hr period. The current is monitored constantly with a Speedomax recorder to facilitate the observation of current fluctuations. The magnetic field is measured by a Varian Associates proton fluxmeter, Model F6.

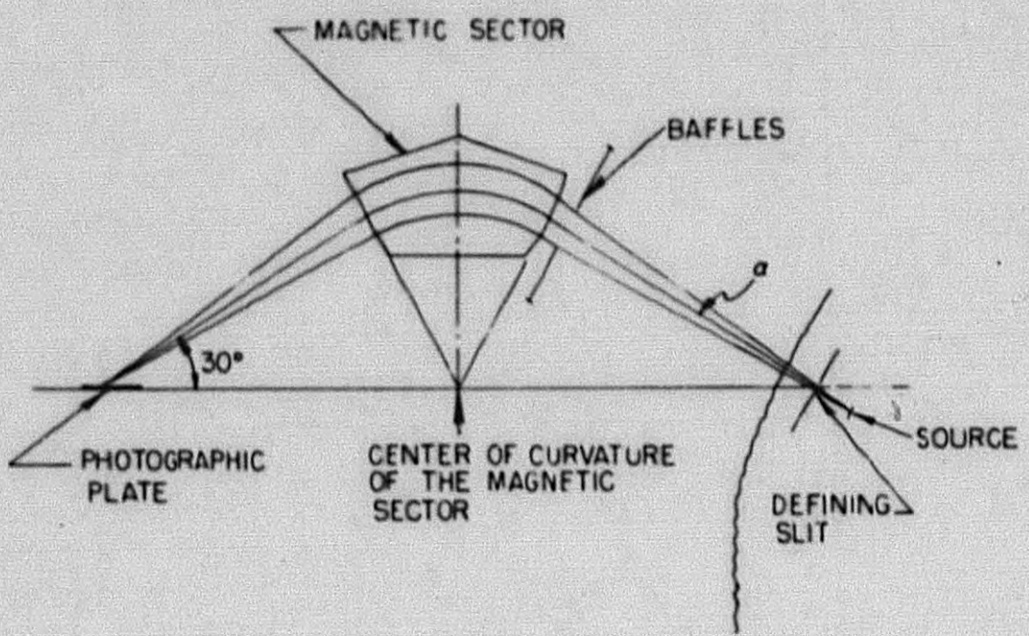

Fig. 1. Schematic diagram of low-geometry spectrograph.

The vacuum in the spectrograph is maintained by two 260-liter/sec oil diffusion pumps each backed by Duo Seal mechanical pumps. The source and receiver units of the spectrograph are housed in separate chambers which can be sealed off from the main vacuum tank. These two chambers have their own mechanical roughing pumps.

The receiver chamber houses a photographic plate holder which carries the plates used for detection of the alpha particles. The photographic plates are 9-by-2-inch Eastman NTA plates with emulsions 25 or 30 microns thick. The plate holder is set at an angle of 30° to the paths of the alpha particles rather than normal to them. The plate holder or receiver has a lighttight camera door which can be operated from outside the spectrograph. The door makes it possible to expose the photographic plate to the beam of alpha particles but yet keep the plate in the dark while transporting it to the darkroom.

The source holder contains a probe on which the radioactive samples are mounted for exposure in the spectrograph. Various combinations of defining slits, varying in width from 1/8 inch to 5 mils, are placed in front of the sample, depending upon the nature of the experiment. For experiments requiring the maximum geometry, 1/8-inch-wide slits are placed 3/8 and 5/8 inch from the source. For experiments requiring better resolution, a 1/16-inch or 18-mil-wide slit is placed 3/8 inch away. Since the resolution of the spectrograph is very greatly dependent on the width of the masking slit, a description of the slit systems employed is given for each experiment as it is discussed later. The different slit arrangements, together with the baffle system located close to the magnet gap, allow a variation in the solid angle from 10^{-4} to 10^{-7} of 4π.

During an experimental run with the alpha-particle spectrograph, a photographic plate is exposed to the beam of alpha particles. Since the plate holder is at a 30° angle to the beam, the alpha particles enter the emulsion at a 30° angle rather than perpendicular to it. Owing to the small angle of acceptance in the vertical direction permitted by the pole-tip gap, the alpha tracks on the plate should be very nearly parallel. The developed emulsions are examined under a 450-power microscope

with bright-field illumination. The alpha tracks are counted individually, and only those tracks which are parallel are counted. This allows a partial discrimination against background tracks, of which many originate from the inside surfaces of the main vacuum tank (due to radioactive recoil nuclei emitted from the sample) and in general would not strike the photographic plate in a horizontal direction. The track length for a 6-Mev alpha particle is about 25 microns.

In the plate-scanning process, the tracks are totaled for a vertical scan across the plate. The microscope field scans a 1/4-mm-wide strip. A complete counting would consist of making scans every 1/4 mm across the length of the plate. The scan counts are plotted against the position of the scan in the horizontal direction (which can be calibrated in terms of energy) to give the observed spectrum. Only in a few cases is a complete scanning job done. Very often a 1/4-mm-wide scan is taken only at each millimeter position on the plate. When the resolution of the alpha groups is very good, counts are made at every 1/4-mm position over the peaks. For strong lines where reading a complete vertical scan would be an extremely tedious job, only one field of view or fraction of a field is counted for each position (one field of view is about 1/145 of a complete scan).

One of the most important considerations in the use of the alpha-particle spectrograph as a precision instrument is the preparation of the source. Poor samples can cause a lot of energy degradation and result in a prominent low-energy tail and a poor peak half width. To take advantage of the inherently good resolution of the spectrograph, the effects due to poor samples must be held to a minimum. The ideal sample would be a uniformly thin deposit of the radioactive substance. In general, samples made by evaporating a solution of the material to dryness do not meet the above requirements. Two techniques have been developed, however, which consistently give uniformly thin deposits. These are vacuum sublimation and electrodeposition.

The technique used most often for the preparation of samples in this work was vacuum sublimation. It amounts to evaporating a solution of the material (usually a chloride or nitrate of the radioactive species)

on a tungsten filament. A plate (generally 2-mil platinum or aluminum) is held a short distance above the filament (generally about 1/8 inch away). This arrangement is evacuated to a pressure of less than a micron of mercury, then the filament is flashed to white heat by passing current through it to vaporize the material onto the collecting plate. Proper formation of the fil ment into a "U" shape (by heating it in a specially prepared die) permits the material to be well defined on the collecting plate without use of a masking slit between it and the filament. This eliminates the problem of lining up the slit and filament. Depending on how far away the collecting plate is from the filament, the samples made in this manner generally cover a rectangle of about 1/8 by 1 inch. Variations in the vaporizer geometry are made according to the amount of material available and the nature of the experiment. When the best possible resolution is desired, a lower yield is accepted in order to get a more uniformly thin sample. Vaporizer yields usually vary from 50% to 95%, depending on the conditions used. Generally, samples made with this technique give satisfactory spectra when exposed in the spectrograph. For extremely good resolution (peak half widths of about 7 or 8 kev), samples of less than 5 micrograms of material are needed. Under no conditions has a sample of over 100μg been used.

The electrodeposition technique mentioned before has not in general resulted in samples as good as those obtained by vacuum sublimation. Usually other material besides the metal or hydroxide of the radioactive species plates out, causing sample thickness. However, improved techniques for plating the actinides, which have eliminated the inert deposits, have been developed by Harvey.[8] If the electrolysis cell is properly shaped, a well-defined sample along a narrow line can be deposited.

The important considerations in determining how large a sample one should use are the specific activity of the species, the spectrograph geometry, and the nature of the experiment. Combining the spectrograph geometry of 0.004% with the 100μg sample limit makes a study of a radioactive species of half life longer than 10^6 years unfeasible. Very few studies were made of isotopes having half-lives longer than 10^4 years.

The resolution of the spectrograph is inherently good. To a first approximation, the width of the image on the photographic plate is twice the defining slit width at the source. For very small slit widths, however, sample effects make it extremely difficult to achieve this condition.

In experiments dealing with weak sources or sources of materials of low specific activities, the machine background becomes important. In the course of the alpha-emission process, the daughter nuclei recoil with about 100 kev energy. If they recoil away from the sample into the spectrograph, they eventually build up a coating on the walls of the spectrograph, leading to a general alpha contamination of the inside. Although many of the alpha particles emitted by these recoil nuclei do not reach the photographic plate, and of those that do many enter at angles away from the horizontal, an appreciable number of background tracks are counted in the scanning process. The peak-to-background ratio assumes a large importance in low-intensity groups. For this reason, the spectrograph insides have been shielded with aluminum sheeting, which collects the recoils. Periodically the spectrograph is disassembled and new sheeting installed to reduce the machine background.

The energy dispersion at the receiver is given by

$$\text{dispersion} = \frac{E_0}{2r_0},$$

where r_0 is the radius of the normal trajectory and E_0 is the energy of the normal particle. The magnetic field B (in gauss) is related to r_0 (in centimeters) and E_0 (in electron volts) by

$$B = \frac{144}{r_0} E_0^{1/2}.$$

Since r_0 has not been determined exactly (it is about 75 cm), the following equation obtained from the two above is best used for the dispersion:

$$\text{dispersion} = \frac{r_0 B^2}{41{,}472}$$

Thus, if the dispersion is measured for a given field strength, an effective r_0 can be calculated. This could be used to calculate the dispersion for other field strengths. In practice, the dispersion is calculated for an r_0 of 75 cm, and this multiplied by a correction factor (determined experimentally) to yield the correct dispersion. The correction factor is obtained by running samples containing more than one well-known alpha group to determine the dispersion for a particular field strength. This is then compared with the calculated dispersion. Many determinations of this nature have been made, indicating that the actual dispersion is larger than the calculated dispersion by a fairly constant percentage. The difference varies with the alignment of the instrument, and the spectrograph must be recalibrated after every disassemblage. Correction factors from 4.6 to 5.7% were obtained for the various stages of the current work. Asaro[9] has previously shown the constancy of the dispersion over the 9-inch horizontal direction of the photographic plate.

After an exposure has been counted and plotted, the positions of the middles of the peaks at half maximum intensity are determined in order to find the separation between peaks. This was shown to be less sensitive to the location of the peaks on the plate than use of the high-energy edges of the peaks to measure separations. A fair degree of high-energy tailing is evident for all peaks near the low-energy edge of the plate, so, if possible, the groups of interest were generally placed near the middle or on the high-energy half.

The newer high-geometry alpha-particle spectrograph is somewhat more versatile than the lower-geometry spectrograph just discussed. The use of a 180° double-focusing magnet allows geometries of up to 0.1%, depending upon the angle of acceptance of the beam. The pole tips have been carefully designed to permit a resolution comparable with that of the low-geometry spectrograph.

Most of the operating features of the double-focusing alpha-particle spectrograph (Fig. 2) are the same as those of the low-geometry unit. The magnet current supply and vacuum systems have been designed to give the same performance as those of the older unit. There are some additional features, however, that should be mentioned.

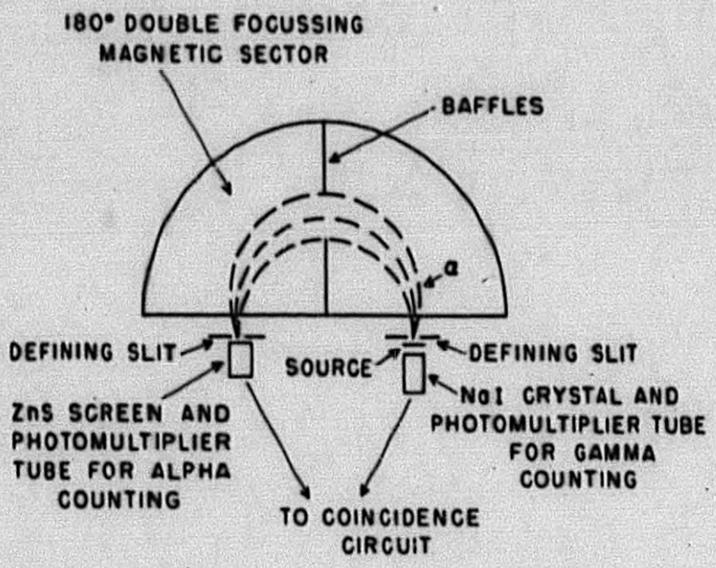

Fig. 2. Schematic diagram of high-geometry spectrograph setup for alpha particle–gamma ray coincidence experiment.

Because of its fairly high geometry and good resolution this spectrograph has been designed specifically to aid in studies of alpha particle-gamma ray coincidences (where coincidences with a particular alpha group are desired). For this purpose, a special receiver unit consisting of a holder for a photomultiplier tube with a lucite window coated with ZnS has been installed. An adjustable slit is placed in front of the ZnS screen. The magnetic field is varied to focus a particular alpha group on the slit, and the scintillations caused by these alphas can be counted and fed into a coincidence unit to serve as gate pulses. The source holder has been constructed with a beryllium window to permit the gamma rays from the sample to strike a gamma scintillator outside the spectrograph. Coincidences can then be run between the particular alpha group gates and the gamma spectrum. This setup should be a great aid in the study of decay schemes of alpha emitters having a large number of alpha transitions. Additional information obtainable with this setup is the angular correlation between specific alpha groups and specific gamma rays, as the gamma-counting device can be set at any desired angle with respect to the source.

The special coincidence receiver unit can be replaced by a photographic plate holder and the spectrograph operated like the older instrument. The plates for this holder measure 1 by 9 inches. So far, the general use of the new spectrograph has been restricted to alpha particle-gamma ray coincidence studies.

B. Gamma-Ray and Coincidence Equipment

The equipment used for the study of gamma-ray spectra and for coincidence measurements has been recently described completely by F.S. Stephens[10] in conjunction with his scintillation spectroscopy studies. Only a general discussion of the main features is given here. The measurements carried out along this line in the course of this work were limited to raw gamma-spectrum determinations, gamma ray-gamma ray coincidence studies, alpha particle-gamma ray coincidence studies, and specific alpha group-gamma ray coincidence studies.

The raw gamma-ray spectra were observed with a thallium-activated sodium iodide crystal as a gamma-ray detector. The fluorescence output from the crystal was converted into an electronic pulse in a photomultiplier tube. This pulse was amplified and fed into a 50-channel pulse-height analyzer, where the energy spectrum of the gamma rays was recorded.

Various corrections must be applied to the data taken in this way. The main corrections are those for the escape peak and for counting efficiency. Axel[11] has made calculations of the magnitude of the escape-peak effect as a function of geometry. These seem to be in good agreement with experimentally observed values, and they have been used in making the necessary corrections to the work described herein. The counting efficiency corrections that have been made are those suggested by Kalkstein and Hollander[12] in a review of the subject.

The circuit used for the various coincidence studies has a resolving time of about 3×10^{-6} second. The gate pulses are passed through a single-channel analyzer in which a discriminating circuit allows the selection of pulses of any particular height. This allows a certain amount of energy selection on the gate side. The signals in coincidence with the gate pulses are recorded by the 50-channel analyzer.

For gamma ray-gamma ray coincidence studies, NaI crystals were used to detect both the gate and signal gamma rays. For coincidences involving the entire alpha-particle spectrum, a ZnS screen coated on an RCA 5819 photomultiplier tube served as the gate alpha detector. The setup used for the specific alpha group-gamma ray coincidence experiments has already been described.

III. EXPERIMENTAL RESULTS

A. Alpha Decay of Americium-243

The interest in this particular nuclide stems from several points: (a) the early work[13] on the decay scheme of Am^{243} showed it to be quite similar to that of Am^{241}, (b) it provides an opportunity to compare the alpha-decay patterns of two odd-mass nuclides of the same element, and (c) it is of interest to gather accurate data on alpha-particle energies and abundances in nuclides suspected of showing rotational band patterns, in order to test quantitatively the Bohr-Mottelson theory.

The earlier work referred to above was that of Asaro and Perlman,[13] in this laboratory, who investigated the alpha and gamma spectra of americium samples enriched in Am^{243}. In their alpha-particle studies, only 2% of the alpha activity in their americium was due to Am^{243}. They were able to assign to Am^{243} three alpha groups having the following energies (in Mev) and abundances: 5.267 (84%), 5.225 (13%), and 5.171 (approximately 3%). A further enriched sample having twice as much Am^{243} activity as Am^{241} was used for the gamma-ray analysis. This showed a very prominent 75-kev gamma ray in coincidence with 80% of the total Am^{243} alpha particles. This 75-kev gamma ray is most certainly an E1 transition (from conversion coefficient). This information led them to propose a decay scheme for Am^{243} in which the main alpha group populates a level 75 kev above the ground state of Np^{239}. This is very similar to the Am^{241} decay, in which the main alpha group populates a level, 60 kev above the ground state, which de-excites by a very prominent 60-kev E1 transition.[13]

A further similarity was noted between the two decay patterns in the energy separations and abundances of the main alpha groups. Am^{241} was reported then to have five alpha groups of energies and abundances as follows:[13] 5.535 (0.3%), 5.503 (0.2%), 5.476 (84%), 5.433 (13.6%), and 5.379 (1.4%). The three lowest-energy groups are separated by almost the same energies and have nearly the same abundances as the three groups reported for Am^{243}. In both cases these three groups were interpreted as populating a Bohr-Mottelson rotational band whose levels follow an I(I+1)

pattern energywise. Knowing the energy separations of three of the levels belonging to the rotational band, one can calculate the spin of the lowest level (I_0). In both cases this was 5/2. In Am^{241} a 5/2 spin assignment for the 60-kev level is consistent with the decay by an E1 transition to the ground state of Np^{237}, which has a measured spin of 5/2.[14] However, for the Am^{243}, there is an inconsistency, since the ground state of Np^{239} has been determined to be 1/2.[15] Because the spin calculations for the 75-kev level are very sensitive to small energy changes, it was deemed of value to determine as accurately as possible the energy separations of the alpha groups believed to be populating a rotational band.

Further, one notes that the ground-state alpha transition in Am^{243} decay was not observed by Asaro and Perlman. Their limit of detection was 2%, and one might expect the transition to be present in much lower abundance than this, judging from its abundance in Am^{241} decay.

Two different samples of americium isotopes were available for the studies here reported. They were both produced by intensive neutron irradiation of Pu^{239} in the MTR reactor. The reactions producing the americium isotopes are as follows:

$$Pu^{239}(n,\gamma)\ Pu^{240}(n,\gamma)\ Pu^{241}(n,\gamma)\ Pu^{242}(n,\gamma)\ Pu^{243}$$
$$\downarrow\beta^- \qquad\qquad \downarrow\beta^-$$
$$Am^{241}(n,\gamma)\ Am^{242}(n,\gamma)\ Am^{243}$$

The first sample used in this study had an activity ratio, Am^{243} to Am^{241}, of about 1 to 4. The activity ratio of the second sample was about 6 to 1 (Am^{243} to Am^{241}). The americium was chemically separated from the fission products and other actinides in both cases. In the second sample, there was some Pu^{238} contamination (having an alpha-particle energy very close to that of Am^{241}); it was present in the same order of magnitude as the Am^{241} (activitywise).

The samples used for exposure in the alpha-particle spectrograph were prepared by vacuum sublimation in a manner already described. The total alpha activities of the two americium samples were 4.6×10^7 d/m for the sample of low Am^{243} content and 1.8×10^7 d/m for the sample of

high Am^{243} content. The spectrograph exposure times in this series of experiments varied from 15 hours to 5 days.

1. Alpha-Particle Energy of Americium-243

Before the fine structure of this alpha decay is considered, the energy determination of the main group is discussed. The first sample available for this series of experiments contained fairly comparable amounts of Am^{241} and Am^{243} with no Pu^{238} impurity. Because the alpha-particle spectrum of Am^{241} has been carefully studied by Asaro,[16] the main alpha group of Am^{241} can be used as a reliable standard for the determination of the energy of the main group of Am^{243}. Asaro and Perlman[13] have reported 5.267 Mev as the energy of the main alpha group of Am^{243}. This value is based on a single determination. Since this run was made, the spectrograph dispersion has been recalibrated, and it appears that their value should be 5.268 Mev. We have continued this determination, using the same alpha-particle spectrograph. The results are summarized in Table I. Experiment 272 is that previously reported by Asaro and Perlman and changed to conform to the newer calibration values. Experiments 300 and 302 were performed with the sample that contained about 20% Am^{243} (activitywise). All three of these determinations were made with 5.476 Mev as the energy of the Am^{241} a_{60} standard.[13, 16] The average value of 5.266 Mev is probably good to 5 kev, considering the uncertainties in the Am^{241} energy (± 2 kev), the location of the peaks on the photographic plate (± 1 kev), and the dispersion calibration of the spectrograph (± 1%). The energies of other Am^{243} alpha groups will be determined relative to the main 5.266-Mev group.

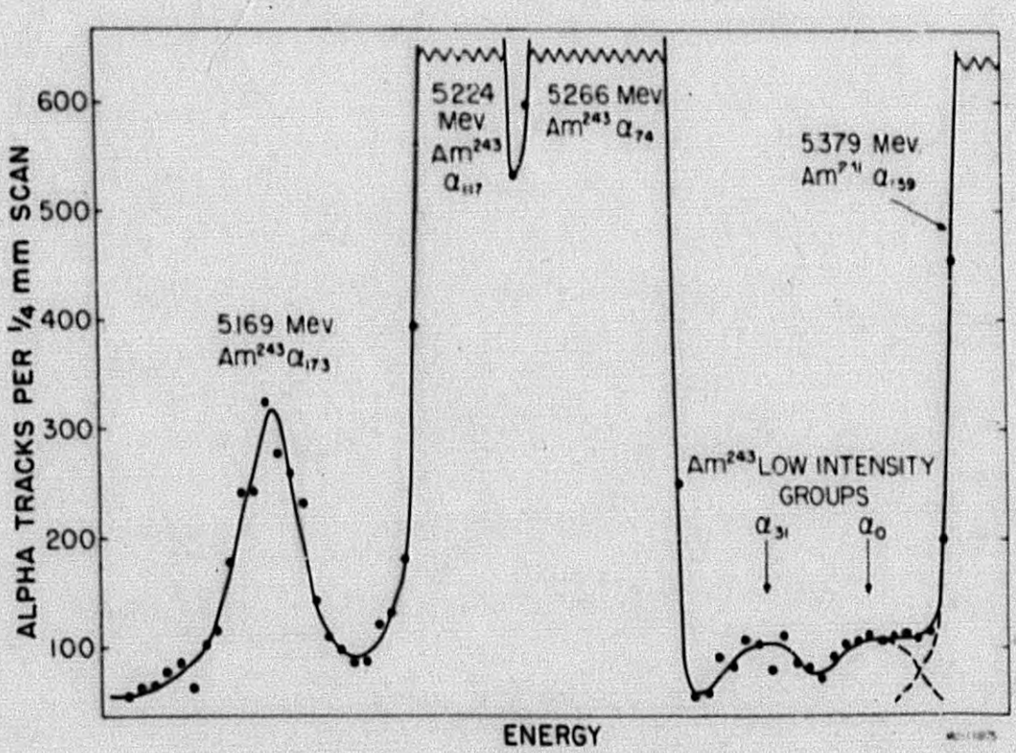

Fig. 3. Alpha-particle spectrum of americium sample of high Am^{241} content.

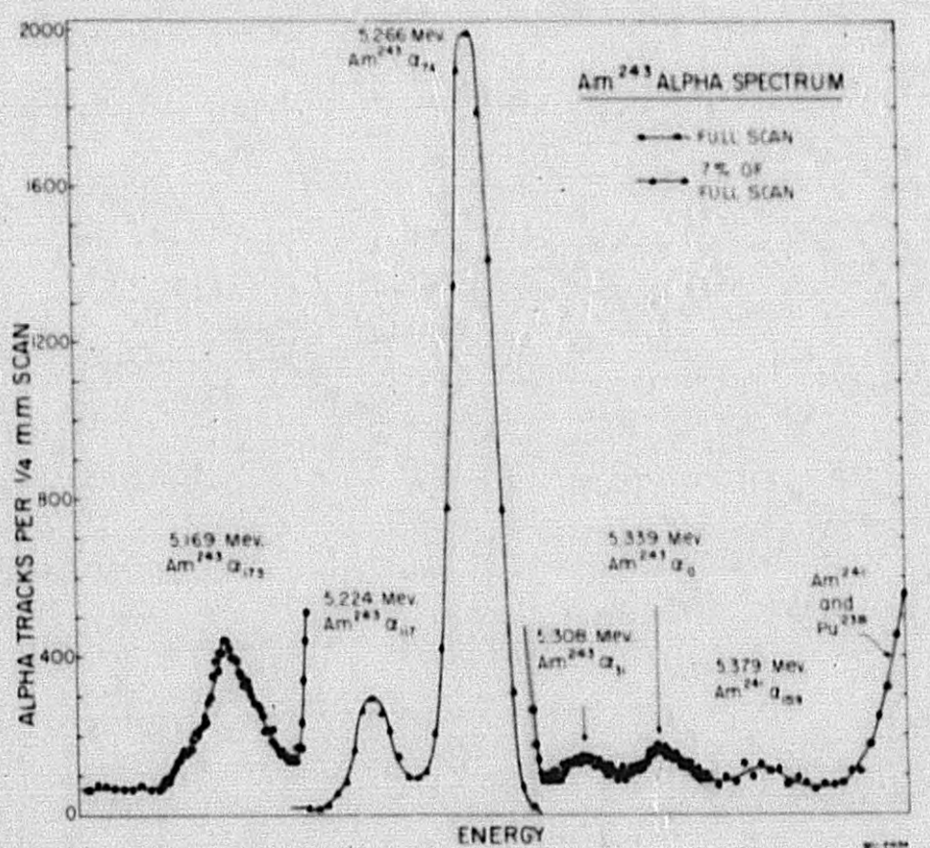

Fig. 4. Alpha-particle spectrum of americium sample of high Am^{243} content.

makes it necessary to operate the spectrograph at maximum transmission. This limit for Am^{243} is such that the spectrograph exposures had to be made with only the 1/8-inch-wide slit system masking the sample in order to see the low-intensity groups with reasonable exposure times.

Table II summarizes the data concerning the two low-intensity groups. Experiment 309 is the run made with the first sample. Experiments 327 and 331 were run with the second sample.

Table II

Low-intensity high-energy alpha groups of Am^{243}

Experiment No.	Energy separations		Abundance ratios	
	$a_0 - a_{74}$ (kev)	$a_{31} - a_{74}$ (kev)	a_0/a_{74} ($\times 10^3$)	a_{31}/a_{74} ($\times 10^3$)
309	73.3	40.2	---	---
327	73.8	44.1	1.90	1.83
331	72.0	41.9	2.06	1.85
Best values	73.0	42.4	1.98	1.84

When corrections are made for the recoil energy, the decay-energy separations from a_{74} become 74.2 kev for a_0 and 43.1 kev for a_{31}. On the basis of the data of Asaro and Perlman, where a 75-kev gamma ray follows most of the alpha emission,[13] the alpha group 73 kev higher in energy than the main group is taken as the ground-state transition. Thus the main group has been designated as a_{74}, as it is separated from the group assumed as a_0 by 74 kev decay energy. The energies of a_0 and a_{31} are 5.339 Mev and 5.308 Mev, respectively.

Because of the tailing contribution of the Am^{241} in this region, no effort was made to integrate the peaks observed in Experiment 309. However, one can arrive at a good estimate for the abundances of the low-

intensity groups by considering the peak heights. This method indicates an abundance relative to α_{74} of 2.3×10^{-3} for both low-intensity groups, in good agreement with the abundances relative to α_{74} obtained from Runs 327 and 331 by integrating the peaks. The assignment of these two groups to Am^{243} is based on their constant abundance, relative to α_{74} of Am^{243}, over a large change in the isotopic constitution of the americium samples studied.

3. Main Alpha Groups of Americium-243

The remainder of the alpha spectrum of Am^{243} is essentially the same as that reported by Asaro and Perlman.[13] A number of shorter runs, each on the order of a day in length, were made to determine the energy separation and abundance ratio of the two most abundant groups. All eight of these runs were made with an 18-mil defining slit in front of the sample, so that the half widths of the peaks were generally about 8 or 10 kev. A typical spectrum run at high resolution is shown in Fig. 5. A summary of the data from these eight exposures is given in Table III, where the exposure number, energy separation of the two main alpha groups, and abundance ratio are given. Exposures 304 and 307 were made with the 20% Am^{243} sample, and the other exposures were made with the larger-percentage Am^{243} sample. In all cases the peaks were integrated to determine the abundance ratio. After 0.7 kev is added to correct for the recoil energy, the decay-energy difference between the levels populated by the two most abundant groups is 43.2 kev. Since the most abundant group at 5.266 Mev has been denoted as α_{74}, the lower-energy group 42.5 kev away will be called α_{117}, as it populates a level 117 kev above the ground state. The energy of α_{117} then becomes 5.223 Mev.

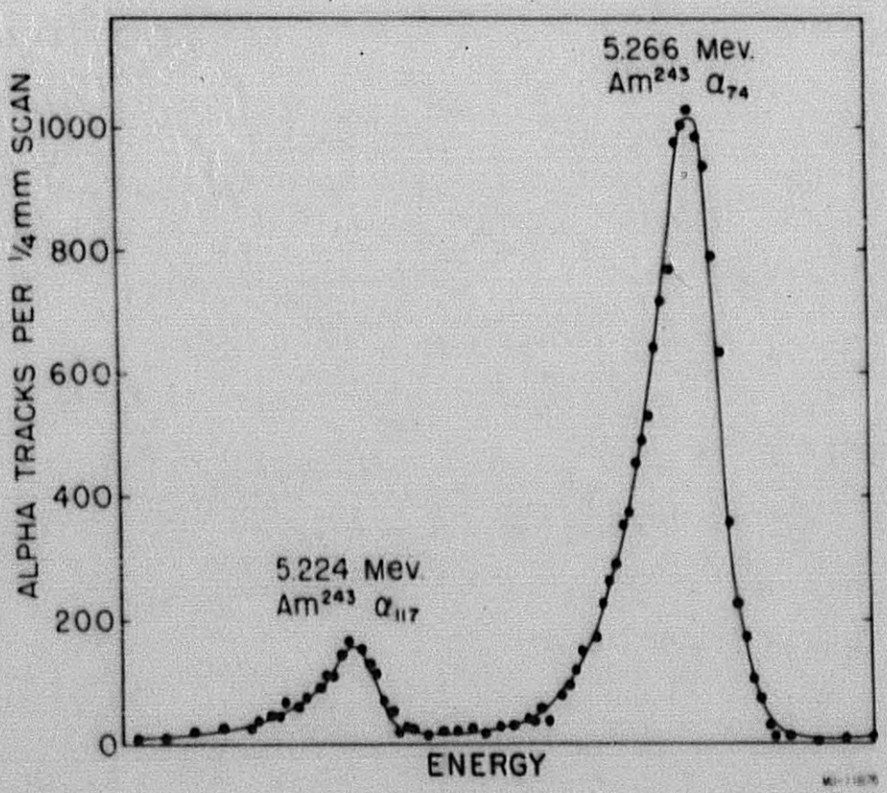

Fig. 5. High-resolution alpha-particle spectrum of Am^{243} main alpha groups.

Table III

Main alpha groups of Am^{243}

Exposure number	a_{117}-a_{74} energy separation (kev)	a_{117}/a_{74} abundance ratio
304	42.6	0.130
307	42.3	0.137
363	42.2	0.127
364	42.4	0.138
367	42.6	0.128
368	42.7	0.132
369	42.5	0.131
370	42.6	0.134
Best values	42.5	0.132

A few words about the limits of error involved here are in order. The average deviation on the energy separation is about 0.2 kev, which may be taken as the uncertainty in locating the peaks on the photographic plate. To this must be added the uncertainty in the dispersion calibration, which is at most 1% or 0.4 kev. The energy separation between these two groups would then be 42.5 ± 0.6 kev. Concerning the uncertainty in the abundance ratio, the effects to be considered are not as clear as for the energy-separation determination. The average deviation in this case is 0.003. The standard statistical deviation for each run, determined from the number of tracks recorded and from a 3% counting error,[17] is about 5.5%. For the eight runs taken together, this becomes about 2%. Therefore, the average deviation of 0.003 seems to be a fair limit of error. The abundance ratio is thus taken to be 0.132 ± 0.003.

The same samples were subjected to longer exposures on the alpha-particle spectrograph to get a statistically significant number of tracks in the previously reported approximately 3% group. These exposures

were generally about 5 days in length. (The results from these runs are given in Tables IV and V.)

The energy separations of this last group from the main groups are given in Table IV. Where the separation from the main group (α_{74}) was measured, the location of α_{74} was determined by taking only a partial scanning in that region of the photographic plate. A full field of view or a fraction thereof was counted at three different vertical positions on the plate and totaled to give a count for that particular horizontal position. The three vertical positions were at the center and the middles of the top and bottom halves. These partial-scan counts were then plotted against the horizontal position on the plate in the usual manner. This procedure was used because the lines on the plates were observed to be somewhat curved and not exactly vertically straight lines. It is felt that counting in the three positions and adding up the results gives a fairly good average peak position with a minimum of counting effort. Where the separation from α_{117} was measured, both peaks were scanned completely in the counting process. The numbers in the α_{173}-α_{117} column in parentheses are the separations between the ~3% group and α_{117}, which were calculated from the measured α_{173}-α_{74} separations and the 42.5-kev separation of α_{117} and α_{74}.

A few more comments on Table IV are in order. Note that two of the five experiments listed were run at high resolution with an 18-mil defining slit in front of the sample (see Fig. 6), and that the other three were run at a higher-geometry arrangement (lower resolution) of a 1/8-inch slit system (Figs. 3 and 4). The exposures taken at high resolution would be expected to give the best energy measurements because of the smaller peak half widths and accompanying smaller uncertainty in peak positions. Because the spectrograph was disassembled for decontamination and moving between the times that Experiments 371 and 434 were run, a new dispersion calibration was needed for Run 434. This was gotten by using the α_{117}-α_{74} separation to define the dispersion (α_{74} was counted only partially). These considerations lead one to conclude that Run 371 represents the best individual experiment on the α_{173}-α_{117} separation. The plate from Experiment 309

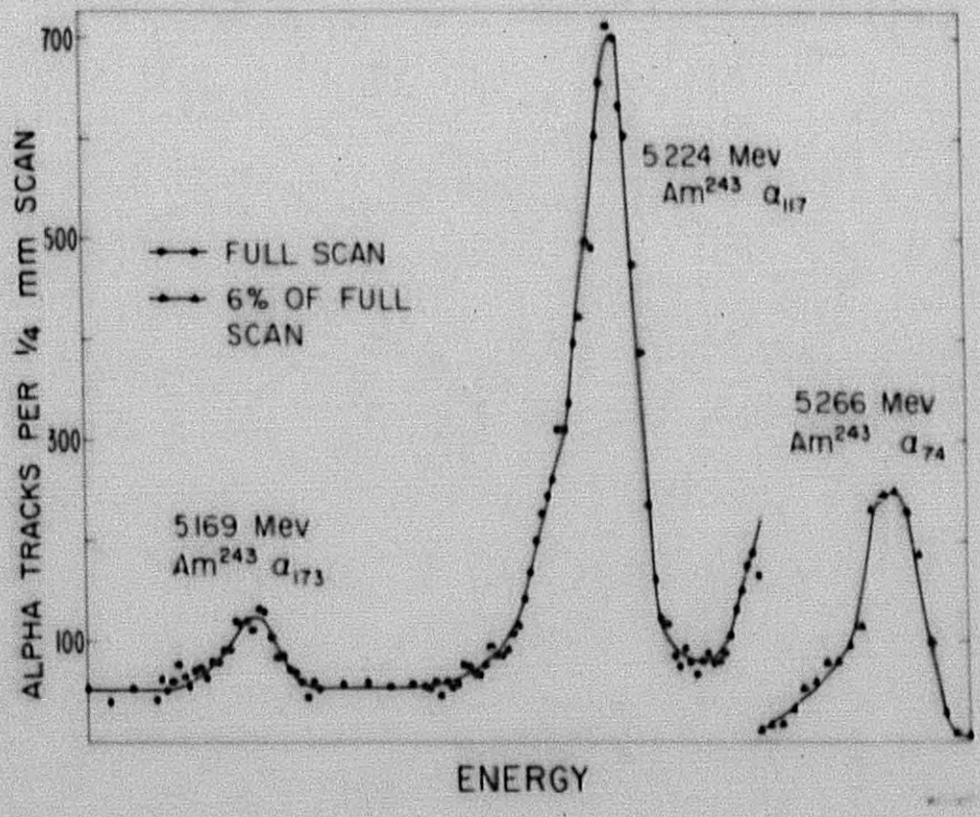

Fig. 6. Main components of Am^{243} alpha-particle spectrum shown at high resolution.

was scanned only at every millimeter (rather than every 1/4 mm); thus, one would not weigh it as heavily as the other runs, where all the scanning was done every 1/4 mm. The average deviation (±0.4 kev) is taken as the uncertainty in the locations of the peaks.

Table IV

Energy determination of the third most abundant alpha group of Am^{243}

Experiment	Slit width	α_{173}-α_{74} separation (kev)	α_{173}-α_{117} separation (kev)
309	1/8 inch	97.7	(55.2)
327	1/8 inch	97.2	(54.7)
331	1/8 inch	98.1	(55.6)
371	18 mil	----	54.4
434	18 mil	----	54.6
		Best value	54.8

Again a 1% dispersion calibration uncertainty is included, giving a value of 54.8 ± 0.9 kev for the separation of the low-energy group from α_{117}.

Corrected for the recoil energy, the decay-energy separation of the low-energy group from α_{117} is 55.7 kev. The decay-energy separation from α_0 is 173 kev, thus the group is called α_{173}. With 5.266 Mev as the energy of α_{74}, the particle energy of α_{173} is 5.169 Mev.

The exposures listed in Table IV also give data on the abundance of α_{173}, which was previously reported as approximately 3%. Table V summarizes the available data regarding the abundance of α_{173}. Because exposure 309 was scanned only every millimeter, the peak integration was not carried out to determine the relative abundances. However, a rough comparison, using peak heights, gives an α_{173}/α_{74} abundance

ratio of 1.3×10^{-2}, in good agreement with the more accurate data in Table V. Where the $\alpha_{173}/\alpha_{117}$ ratio was determined (Runs 371 and 434), both peaks were counted completely.

Table V

Intensity of Am^{243} α_{173}

Experiment No.	Abundance Ratios	
	$\alpha_{173}/\alpha_{117}$	α_{173}/α_{74} ($\times 10^2$)
327	-----	1.59
331	-----	1.49
371	0.104	(1.37)
434	0.131	(1.73)
	Best value	1.54

The α_{173}/α_{74} ratio was then calculated by use of the value of 0.132 for the α_{117}/α_{74} ratio (Table III). In the other two cases, the α_{173} peak was counted completely and the α_{74} peak total was calculated from the total α_{74} tracks recorded on a shorter exposure. The uncertainty in the α_{173}/α_{74} abundance ratio is taken as the average of the runs listed in Table V (± 0.14).

Table VI

Complex alpha decay of Am^{243}

Group	Particle energy (Mev)	Abundance relative to α_{74}	Abundance (%)
α_0	5.339	1.98×10^{-3}	0.17
α_{31}	5.308	1.84×10^{-3}	0.16
α_{74}	5.266	1	86.9 ± 0.4
α_{117}	5.224	0.132	11.5 ± 0.3
α_{173}	5.169	1.54×10^{-2}	1.3 ± 0.2

Table VII

Complex alpha decay of Am^{241} (Asaro and Perlman)

Group	Particle Energy (Mev)	Abundance (%)
α_0	5.535	0.3
α_{33}	5.503	0.2
α_{60}	5.476	84
α_{103}	5.433	13.6
α_{159}	5.379	1.4

The energies and abundances of the five alpha groups observed in Am^{243} decay are summarized in Table VI. The alpha spectrum of Am^{241} is given in Table VII for comparison.[13]

4. Half Life of Americium-243

The first sample of americium available for this study contained large amounts of both Am^{241} and Am^{243} with no interfering Pu^{238} contamination. If one knows the mass and activity ratios of Am^{243} and Am^{241} in the sample, the following relationship can be used to calculate the half life of Am^{243}:

$$T_{1/2}(243) = T_{1/2}(241) \frac{(N_{243})}{(N_{241})} \frac{(A_{241})}{(A_{243})},$$

where $T_{1/2}(241)$ is 461.3 years,[18] (N_{243}/N_{241}) is the mass ratio, and (A_{241}/A_{243}) the activity ratio. This relationship is readily derived from the simple radioactive decay law. Since the mass ratio is readily measured with a mass spectrograph, the half-life determination becomes a matter of determining the activity ratio. This can be done with the aid of the alpha-particle spectrograph.

The sample referred to above was mass-analyzed by Frederick L. Reynolds, and a mass ratio (243/241) of 4.27 ± 0.06 was reported. Earlier data taken by Frank Asaro, using the same spectrograph as used in this series of experiments, are also considered and discussed in the light of the new results on the complex alpha spectrum of Am^{243} reported herein. The sample of americium in Asaro's work had a mass ratio (243/241) of 0.340.

The activity ratio can be determined two different ways with the spectrograph. One way would be to put both sets of peaks (Am^{241} and Am^{243}) on the same photographic plate by proper adjustment of the field (they would be separated by about 60 mm). A second approach would be to use two exposures. In the first exposure the Am^{243} peaks can be placed at a definite position on the plate (preferably near the middle). For the second exposure the field would be adjusted to put the Am^{241} peaks in that same position. The two runs would have to be normalized to the same length of exposure. The second method assures the same geometry for both the Am^{241} and Am^{243} groups since they appear at the same position on the receiving plate and thus have the same path lengths from the source to the plate. As the photographic plate is placed at a 30^{o} angle to the principal beam, a geometry correction has to be applied for the first case. The higher-energy alpha particles (those of Am^{241}) travel a longer path than the lower-energy ones (Am^{243}). The longer path means more beam divergence. This means that the photographic plate subtends a smaller vertical angle of the beam of an alpha group on the high-energy side of the plate than on the low-energy side. Specifically, this means a lower geometry for the Am^{241} groups than for the Am^{243} groups. This geometry difference can be corrected for by calculating the path lengths for the various groups of particles. The normal beam travels 335 cm from the source to the plate. A particle striking the plate 60 mm to the higher-energy side would have a path length of 340 cm. This means a 1.5% geometry correction. For fine-structure determinations, this correction factor is generally much smaller (the peaks are closer together) and is not used, since it is practically insignificant compared with the errors in track counting.

The experimental data are recorded in Table VIII. (Experiment 217-218 is the one referred to previously as having been performed by Frank Asaro); the rest are runs made in the study here presented. In most cases only the main alpha group in each set of peaks was counted. Thus, the total alpha tracks for each nuclide were calculated from a knowledge of the fine structure. Of the five determinations reported, only one (303-304) was done with the second method outlined above, where both the Am^{241} and Am^{243} alpha particles have the same geometry. The other two experiments listing two exposures (217-218 and 307-308) were not run in this way. The magnetic field was not changed between exposures. The second plate in each case was exposed for a much shorter time, so that the intense Am^{241} peak could be counted on a shorter exposure and normalized to the longer exposure, thus saving time in counting the tracks. The best value of 7600 years is gotten by weighting the various experiments, first according to their average statistical deviations (based on the number of tracks counted), and then weighting 303-304 doubly, since no geometry corrections were required for that experiment.

The uncertainties involved in this determination indicate limits of ± 370 years for the half life. The average deviation in Table VIII, 150 years (2.0%), is taken as the uncertainty in the alpha-track counting. The uncertainty in the mass-ratio measurements is 1.4% (110 years). The half life of Am^{241} is good to within 0.4% (30 years).[18] To this must be added 1.0%(80 years), which is from the uncertainty in the abundances of the main alpha groups of Am^{241} and Am^{243} (since only the main groups were counted in most cases). This gives a total uncertainty of 4.8% (370 years).

This half life for Am^{243} of 7600 ± 370 years is to be compared with the literature values of $\sim 10^4$ years (Street, Ghiorso, and Seaborg),[19] 7.6×10^3 years (Asaro and Perlman),[13] 8800 ± 600 years (Diamond et al.),[20] and 7930 years (Wallman et al.).[21] The 10^4 years value is only approximate and represents the first estimate of this half life. It was obtained from a mass analysis of Am^{243} and the chemical yield of milked Np^{239}. The value reported by Asaro and Perlman represents the

early result from Experiment 217-218 in Table VIII, and has now been re-evaluated in the light of the new abundance data. The value of

Table VIII

Half life of Am^{243}

Experiment number	Mass ratio (243/241)	Activity ratio (241/243)	Am^{243} $T_{1/2}$ (years)	Remarks
217-218	0.340	50.0	7840	Corrected for different geometry
300	4.27	3.82	7530	Corrected for different geometry
302	4.27	3.97	7820	Corrected for different geometry
303-304	4.27	3.84	7570	Peaks in same position
307-308	4.27	3.72	7330	Corrected for different geometry
		Best value	7600 years	

8800 ± 600 years reported by the Argonne National Laboratory group (Diamond et al.) was obtained in much the same manner as the value presented herein. A mixed $Am^{241-243}$ sample was mass-analyzed and the Am^{241}/Am^{243} activity ratio determined by alpha-pulse analysis. Their value for the Am^{243} half life was calculated using 470 years[22] as the half life of Am^{241}. The newer value of 461.3 years[18] used here is considered to be more accurate. Using the lower value for the half life of Am^{241} gives a half life of 8640 years for Am^{243}. As the uncertainty in the new Am^{241} half life is considerably less than that in

the older value used by Diamond et al., the uncertainty in their half life for Am^{243} can be reduced to 440 years. Thus, their value of 8640 ± 440 years seems to be in disagreement with the value of 7600 ± 370 years reported here. The mass ratio in the sample used by the Argonne group was 0.335 (243/241), and is similar to that in the sample used for Experiment 217-218 in Table VIII. This mass ratio represents a very small activity ratio of Am^{243} to Am^{241} (about 0.02), which may be inherently difficult to measure accurately when tailing from the Am^{241} peaks may interfere with the Am^{243} peaks in a pulse analysis. The use of a high-resolution low-dispersion alpha-particle spectrograph (as in this study) essentially eliminates the tailing problem. The value of 7930 years reported by Wallman et al was obtained by measuring the specific activity of americium metal. This value is in good agreement with the value reported here (7600 ± 370 years), and is probably subject to less uncertainty. On the basis of the half life of 7930 years just mentioned and the abundance data in Table VI, partial half lives for each alpha group in Am^{243} decay have been calculated. These are summarized in Table IX.

Table IX

Partial half lives of Am^{243} alpha transitions

Group	Abundance (%)	Partial half life (years)
α_0	0.17	4.7×10^6
α_{31}	0.16	5.0×10^6
α_{74}	86.9	9.13×10^3
α_{117}	11.5	6.90×10^4
α_{173}	1.3	6.1×10^5

5. 75-kev Gamma Ray–L X-Ray Coincidence Study

The only previous study on Am^{243} indicated the presence of a very prominent 75-kev gamma ray.[13] It was shown that the 75-kev gamma ray was in coincidence with 80% of the alpha particles, which places it as de-exciting the level populated by the 86.9% α_{74} group (5.266 Mev), and makes it an E1 transition. This is very similar to the situation in Am^{241} decay, in which a 60-kev E1 gamma ray follows the main alpha group. It is of interest to examine the mode of de-excitation of the levels populated by the lower-energy alpha transitions. To do this, coincidences were run between the prominent 75-kev gamma ray and the rest of the gamma spectrum. The coincidence circuit used in this experiment has recently been described elsewhere[10] and is not discussed here except to note that the spectrum in coincidence with the 75-kev gates was recorded on a 50-channel pulse-height analyzer. A zinc sulfide screen was used as an alpha-particle detector, and a sodium iodide crystal for the gamma detector.

The americium sample that had a 6-to-1 activity ratio (243/241) was used for this study. Since the Np^{239} daughter has gamma radiation in the 75-kev region, it was necessary to remove the Np^{239} from the sample before doing the coincidence experiment. This was accomplished by running the americium that contained neptunium through a Dowex-A1 anion-exchange column in 10 N HCl containing 0.1 N HNO_3 (to keep neptunium in the penta-positive valence state). The americium was not held up by the column and came through in the first column volume of wash. The neptunium was momentarily held up on the column. The americium fraction was evaporated to dryness and then transferred to an aluminum counting plate for the coincidence study. Two different americium samples were purified and then used in these studies.

The geometry was determined by running a sample of Am^{241} of known alpha counting rate. The total gamma spectrum up to 100 kev was recorded on the 50-channel analyzer. Using the value of 0.40 60-kev gamma rays per alpha particle and one L x-ray for each 60-kev gamma ray,[23] one calculates a geometry factor of 41% and an

attenuation factor for the L x-rays of 1.6. This means that the total L x-rays recorded per 75-kev gamma ray in the coincidence runs on Am^{243} had to be multiplied by a factor of 1.6/0.41 = 3.9 to get the actual number of L x-ray coincidences per 75-kev gamma ray.

Before going to the results of the 75-kev gamma ray-L x-ray coincidences on Am^{243}, it might be a good idea to see what coincidence intensity might be expected. Beling, Newton, and Rose have done the analogous experiment with Am^{241} and have reported 6% L x-rays in coincidence with the 60-kev gamma ray.[24] If we accepted the proposed interpretation of the top three levels in Np^{237} as belonging to a Bohr-Mottelson rotational band,[13] we would expect gamma transitions from the 103- to the 60-kev state, from the 159- to the 103-kev state, and a small amount of crossover from the 159- to the 60-kev level. We would not expect to see any decay from the 103- and 159-kev states to levels lower than the 60-kev state. The expected transitions would be M1-E2 mixtures (except for the E2 crossover transition) and would be essentially completely converted in the L and M shells. Decay from the 103-kev state should give 13.6 transitions to the 60-kev level per 100 total alpha transitions, owing to the direct alpha population to that level. The alpha decay to the 159-kev level would contribute two cascade transitions to the 60-kev state per alpha populating it, or 2.8 transitions per 100 Am^{241} alphas (neglecting the expected small amount of crossover transitions direct to the 60-kev state). This gives a total of 16.4 transitions to the 60-kev level per 100 alpha transitions. To estimate the number of L x-ray-60-kev gamma-ray coincidences we would expect to get, the above number would have to be corrected for the conversion in the lower shells (M, N, etc.) and for the Auger effect. For M1 or E2 transitions we expect an L/M ratio of about 3,[25] or 75% of the internal conversion taking place in the L shell. This reduces the coincidence intensity to 12.3%. Smith and Hollander[26] report an $L_I/L_{II}/L_{III}$ ratio of 1/1/1 for the 43.4-kev transition in Am^{241} decay and an $M_I/M_{II}/M_{III}$ ratio of 1/1/1 for the 55.4-kev transition. (The L lines of the 55.4-kev transition were covered by other lines). Since the L-subshell ratios and the M-subshell ratios as a general rule are

the same,[25] the $L_I/L_{II}/L_{III}$ ratio for each transition is 1/1/1. If one uses this ratio and Kinsey's fluorescence yield information,[27] an expected fluorescence yield for L x-rays of 48% is calculated. This cuts the expected L x-ray-60-kev coincidence intensity to 5.9%. This value is in excellent agreement with the observed intensity of 6% mentioned previously.

To check the coincidence equipment, we have rerun the Am^{241} L x-ray-60-kev gamma ray coincidence experiment in connection with the Am^{243} experiment. Our result of 5.8% coincidence intensity for Am^{241} is in good agreement with the English work. We would conclude from this that the two higher levels (the 103- and 159-kev states) do cascade to the 60-kev level in a manner consistent with a Bohr-Mottelson rotational band interpretation for these levels.

In Am^{243} decay, the alpha populations to the two highest levels (117 and 173 kev) are 11.5% and 1.3% respectively. Thus one would expect 14.1 total transition coincidences with the 75-kev gamma ray per 100 alpha transitions if a rotational band pattern were being followed here. If the L/M ratio and fluorescence yield is the same as for Am^{241}, the expected L x-ray-75-kev gamma ray coincidence intensity would be 5.1%. Table X summarizes the coincidence data. Three runs were made on each sample. The delay between purification and the running of the coincidences was considerably longer with the first sample. One notes that the coincidence intensities from Sample 1 are 10% to 15% higher. Possibly the growth of the Np^{239} daughter was beginning to interfere with the experiment. The runs on Sample 2, where the delay period was much shorter, are therefore considered more accurate.

The experimental 4.2% intensity is somewhat lower than the expected intensity (5.1%) just calculated. This can be interpreted as due to a lower L x-ray fluorescense yield than the Am^{241} data indicated. If we assume the same L/M ratio as used previously for the Am^{241} decay, the apparent fluorescence yield for Am^{243} is 40%. This decrease could be attributed to a larger percentage of conversion in the L_I shell, giving a larger percentage of M1 character to the transitions cascading to the 74-kev state in Am^{243} decay than for the analogous transitions in Am^{241} decay. Smith and Hollander have proposed an 85% M1-15% E2 mixture

for the 43-kev transition in Am^{241} decay to explain the observed L-subshell ratio.[26] This mixture for Am^{243} would then have an even greater percentage of M1 character.

Because of the problem associated with the growth of the Np^{239} daughter, it was not considered feasible at this time to further investigate the gamma rays accompanying the Am^{243} alpha decay.

Table X

Am^{243} L x-ray-75-kev gamma ray coincidence study

Sample	Time after purification (minutes)		L x-rays/75-kev gamma ray
1	220		0.046
	230		0.048
	275		0.043
		average	0.046
2	53		0.038
	63		0.042
	71		0.041
		average	0.040

Best value = 4.2% coincidence intensity

6. Decay Scheme of Americium-243

A decay scheme for Am^{243} is shown in Fig. 7. It is essentially the same as one that appeared in earlier publications[10, 28] from this laboratory, except for minor changes in energies and abundances.

The spins of 5/2 for Am^{243} and 1/2 for Np^{239} are the results of spectroscopic measurements.[29, 15] As mentioned previously, an early

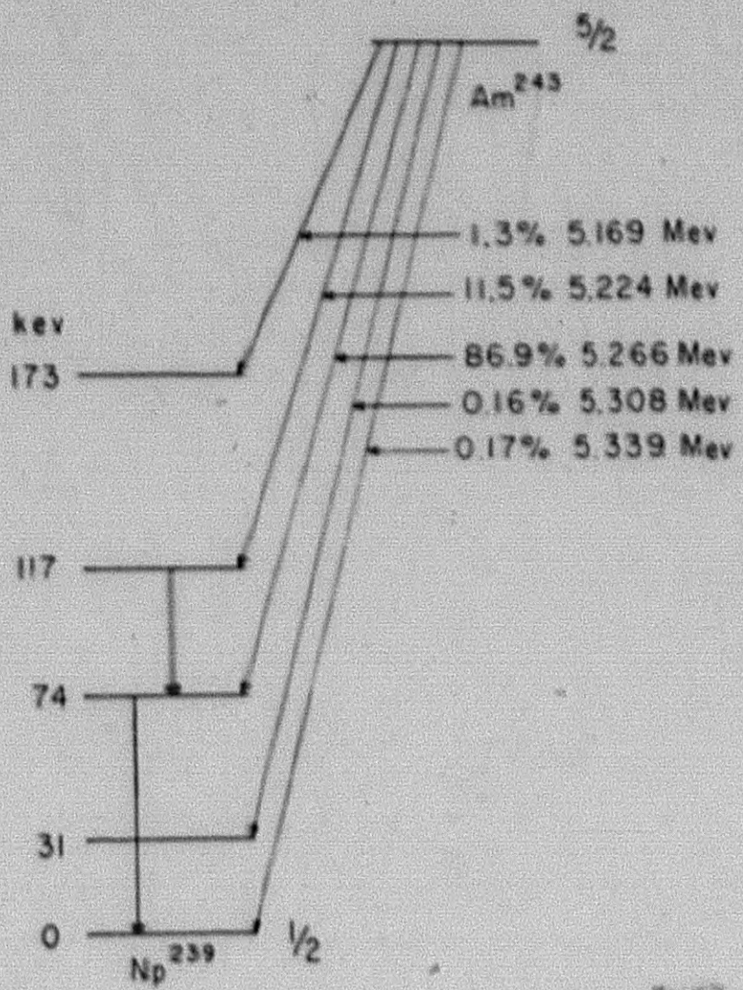

Fig. 7. Decay scheme of Am^{243}.

interpretation of the three highest levels in Np^{239} was that they were members of a Bohr-Mottelson rotational band. This interpretation was based on the regular energy spacings between the levels and the regular decrease in alpha-particle abundances as one goes to the higher levels. The 75-kev gamma ray-L x-ray coincidence measurement just discussed showed that these levels de-excite in a manner consistent with the rotational-band interpretation. Using the I(I+1) dependence for energy, one can calculate the spin of the base member of the band (the 74-kev level). The equations involved here are

$$E_{117} - E_{74} = A(I_0+1)(I_0+2) - A(I_0)(I_0+1),$$

$$E_{173} - E_{117} = A(I_0+2)(I_0+3) - A(I_0+1)(I_0+2),$$

which leads to

$$\frac{E_{173} - E_{117}}{E_{117} - E_{74}} = \frac{I_0 + 2}{I_0 + 1} .$$

where I_0 is the spin of the base level. Previously we determined the decay-energy separations to be 43.2 kev for $E_{117} - E_{74}$ and 55.7 kev for $E_{173} - E_{117}$. With these values, I_0 is 2.46. If one considers the uncertainty in I_0 to be due only to the uncertainty in locating the peaks on the photographic plate, the value of I_0 becomes 2.46 ± .20, clearly indicating a 5/2 spin for the 74-kev level. This is a fair treatment of the uncertainty here, since the calibration error is the same for all runs and is canceled out in the equation above. Thus, one would like to interpret the 74-, 117-, and 173-kev levels as belonging to a rotational band with spins 5/2, 7/2, and 9/2. A discussion of the alpha-decay populations to these levels is reserved for a later section.

It must be mentioned, however, that it is inconsistent with the previous line of reasoning that the 75-kev gamma transition is an E1 transition and decays from the 74-kev level to the ground state, which has a measured spin of 1/2. Furthermore, the alpha-75-kev gamma-ray angular correlation study shows an anisotropy best fit by a $5/2 \rightarrow 3/2 \rightarrow 1/2$ sequence and an $f = 1$ alpha transition.[10,28] This

suggests that the rotational band calculations just cited are not valid. However, one can maintain the rotational band interpretation if one assumes the spin determination of the Np^{239} ground state to be wrong. This is highly speculative, however. Another possibility has been suggested by Rasmussen.[30] He proposes that the 75-kev gamma ray may not terminate at the spin 1/2 ground state but at another level (spin 3/2, 5/2, or 7/2), which is not seen in the alpha-particle work. As yet, there is no proof one way or the other concerning the presence of other levels than those indicated in the alpha-particle spectrum.

It is of interest to note that in Am^{241} alpha decay, where the level separations indicate a 5/2 spin for the 60-kev level, there is no inconsistency in assuming the 5/2 spin for that level. There, the ground state of Np^{237} has a measured 5/2 spin.[14] The 60-kev E1 transition would then be a 5/2 → 5/2 transition. Here also an angular correlation has been observed.[31] It has been shown that this correlation is consistent with a 5/2 → 5/2 → 5/2 sequence if one takes either a p-wave alpha transition or a mixture of s- and d-wave alpha transitions.[32] A similar mixture might also be employed to explain the angular correlation for the Am^{243} case and allow one to keep the rotational-band interpretation for the top three levels.

Recent Coulomb excitation experiments by Newton[33] indicate the presence of a 5/2, 7/2, 9/2 rotational band in Np^{237}, of which the 5/2 level is the ground state, the 7/2 level is at 33 kev, and the 9/2 level is at 75 kev. The 7/2 level at 33 kev is undoubtedly the level populated very weakly by the Am^{241} alpha decay. The 9/2 level at 75 kev is not seen in the alpha decay of Am^{241}. By analogy, one might consider the ground and 31-kev levels in Np^{239} to be members of a similar band. In both nuclides, the alpha population to these states is very small.

Although one can speculate freely about the spins and parities of the states populated by Am^{243} alpha decay, it is very difficult to say much with certainty. Thus, no definite assignments have been indicated in Fig. 7 except the measured spins for the two ground states.

B. Gamma Rays in the Decay of Curium-242

Asaro, Thompson, and Perlman[9] have thoroughly studied the alpha decay of Cm^{242} by making both alpha-particle and gamma-ray measurements. Multipole order assignments have been made for the low-energy gamma rays on the basis of their intensities, which have been substantiated by the conversion-electron study by Smith and Hollander.[34] Thus, the decay scheme of this nuclide is fairly well fixed as far as the low-energy states of Pu^{238} are concerned. In the course of some of the experiments on Cm^{244} conducted in this research (to be described in the next section), a knowledge of the gamma-ray intensities in Cm^{242} was needed. Although these intensities were available from the first work cited above, it was decided that a check on these intensities would be made anyway.

The gamma rays of importance here are the 44-, 100-, and 155-kev transitions. The abundances given by Asaro et al. for these gamma rays are 4.2×10^{-4} (44-kev gamma ray), 6×10^{-5} (100-kev gamma ray), and 2.7×10^{-5} (155-kev transition).

In measuring the intensities of these gamma rays, an accurately alpha-counted sample of Cm^{242} was used. The gamma spectrum from this sample (Fig. 8) was observed with a NaI detector connected to a 50-channel pulse-height analyzer. The various peaks were integrated and the necessary corrections for absorber, counting efficiencies, and escape-peak effects were made in the manner discussed in the section on experimental techniques.

To determine the geometry of the system, an accurately alpha-counted sample of Am^{241} was placed in the same position in front of the counter. Care was taken to make sure that the area covered by both samples was very nearly the same. The observed number of 60-kev photons was then used to determine the geometry factor.[23] This factor was then used to calculate the Cm^{242} abundances.

The abundances of the three gamma rays determined in this way are as follows: 2.9×10^{-4} (44-kev gamma ray), 4.1×10^{-5} (100-kev gamma ray), and 1.8×10^{-5} (155-kev gamma ray). These abundances are all smaller than those given by Asaro. One notes, however, that the

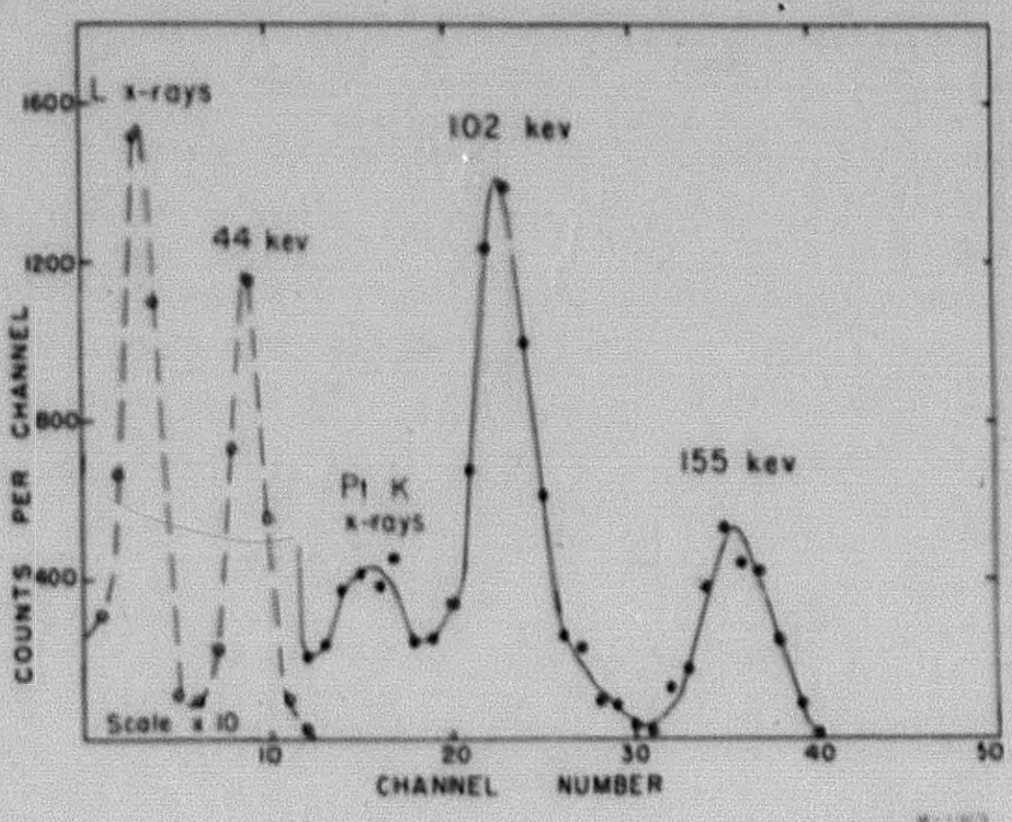

Fig. 8. Gamma-ray spectrum of Cm^{242}.

relative abundances of the three gamma rays are the same in both studies.

These smaller intensities yield somewhat larger conversion coefficients for the 44- and 100-kev gamma rays than given by Asaro et al. However, the previous interpretation of these conversion coefficients in terms of gamma-ray multipolarities is not altered by the new intensities.

C Alpha Decay of Curium-244

A careful study of the alpha decay of Cm^{244} is profitable for a number of reasons. Since Cm^{244} is an even-even nuclide, it is of interest to compare its decay properties with those of other heavy even-even isotopes and note its relationship to the currently expanding systematics for these isotopes. Also, since very accurate data are available on the alpha decay of Cm^{242}, it is of interest to search for similarities and differences between two near-by isotopes of the same atomic number. Another point of interest here, which is elaborated on in the discussion section, is the usefulness of good alpha-transition intensity data in checking a certain corollary of the Bohr-Mottelson collective model.

The first study of the decay scheme of Cm^{244} was by Asaro, Thompson, and Perlman in 1953.[9] They studied the complex alpha spectrum in the alpha-particle spectrograph and reported two alpha groups differing by 42.5 kev and in an abundance ratio of 1 to 1, the highest-energy group being most intense. These observations were made on curium samples containing $Cm^{242,243,244}$ in which the main portion of the alpha activity was due to Cm^{242}. Considering the low level of the Cm^{244} activity in their samples, it would be extremely difficult for them to observe any except the most intense Cm^{244} alpha groups with reasonable exposure times. The rapidly developing systematics of even-even alpha emitters[6] show many cases in the heavy-element region where a group of low intensity, 0.1% or less, appears in the alpha spectrum about 150 kev lower in energy than the main ground-state transition. This is generally considered the alpha transition to the 4+ member of a Bohr-Mottelson rotational band, although in many cases the spin and parity of the level populated in this manner have not been determined. In Cm^{242} alpha decay, the abundance of this alpha group is 0.035%.[9] If such a group exists in the decay of Cm^{244} and if its abundance is comparable with what it is in Cm^{242} decay, it is not at all surprising that the previous study did not show such an alpha transition. With the advent of large amounts of curium in which the Cm^{244} content was high (50% or better, activitywise) it

was possible to reinvestigate the decay scheme of Cm^{244} with a minimum of interference from the radiations of other curium isotopes.

The curium samples used in the study reported here were made by intense neutron irradiation of plutonium in the Materials Testing Reactor at Arco, Idaho. The reactions leading to the curium isotopes of interest are successive neutron captures to give the beta-unstable heavy plutonium isotopes (Pu^{241} and Pu^{243}) that beta-decay to Am^{241} and Am^{243}. Then, neutron capture by these americium isotopes gives the beta-emitting Am^{242} and Am^{244}. Neutron capture by the resulting Cm^{242} and Cm^{244} yields curium isotopes from mass 242 up to 246. The isotopic composition of the curium fraction is dependent on the length of irradiation in the pile. The curium used in the following studies was such that essentially all of the alpha activity was due to Cm^{242} and Cm^{244}. The sample that was used for the alpha-particle studies and some gamma-ray work was about 2/3 Cm^{244} at the start of this work. Later, a sample containing about 98% Cm^{244} was used for some gamma-ray studies. Besides the reactions considered above, additional neutron capture eventually leads to elements above curium. Also, fission reactions yield large amounts of lighter elements. Thus, the chemical separations were designed to separate a curium fraction in a mass-free state from other actinides and fission products.

In searching for very low-intensity alpha groups, lower in energy than very prominent alpha transitions, the source preparation is of utmost importance. Low-energy tailing of the prominent groups may obscure the low-intensity groups. One of the main causes of this tailing effect is the degradation of the energy of the alpha particle by multiple scattering in the source. This effect can be reduced by careful preparation of a uniformly thin source. In the series of experiments here described, several sources of the curium samples were prepared by vacuum sublimation and exposed in the alpha-particle spectrograph. Each source contained about 1μg of material. The region just lower in energy than the two main peaks of Cm^{244} was scanned in the usual manner to determine how bad the tailing effect

was for each sample. Three of these sources were then used for further study. The total alpha-counting rates of the three sources were 6.6×10^8 (A), 4.5×10^8 (B), and 3.8×10^8 (C) alpha disintegrations per minute. Source (C) was the best source as far as the tailing problem was concerned. These three sources were then used for the series of experiments to be described next.

1. Main Alpha Groups of Curium-244

As mentioned previously, Asaro et al.[9] have reported two main alpha transitions for Cm^{244} in an abundance ratio of three to one. In the course of the present study, a series of seven short exposures was made with the three samples to accurately measure the energy separation between the two groups, and their relative intensities. No attempt was made to determine their absolute energies; the energy of the main group is taken as 5.798 Mev , which was the value reported by Asaro et al. in 1953. All these exposures were made with an 18-mil defining slit over the sources (high-resolution condition), so that the peak half widths were generally 7 or 8 kev. A typical spectrum of the main Cm^{244} alpha groups is shown in Fig. 9. The exposure duration for these runs was generally about 10 min, giving about 5000 total alpha tracks in the two peaks. The results from this series of exposures are summarized in Tables XI and XII.

The data concerning the energy separation between the two main groups are given in Table XI. There the particle-energy separation and the decay-energy separation (gamma-ray energy) between the two groups are given for each exposure. Exposure 345 was run with source A; Exposures 347, 348, and 349 with source B; and Exposure 352 with source C. Since the resolution with the 18-mil slit is more than enough to separate completely these two groups, the tailing effect is not an important consideration here. In 348 II and III and 349 II and III, two exposures were made on the same photographic plate. The magnetic field was changed between runs by an amount sufficient to put each exposure on a different region of the plate. That the energy separation did not change much with the positions of the peaks on the plate indicates a fairly constant dispersion over the 9-inch plate. The decay-energy

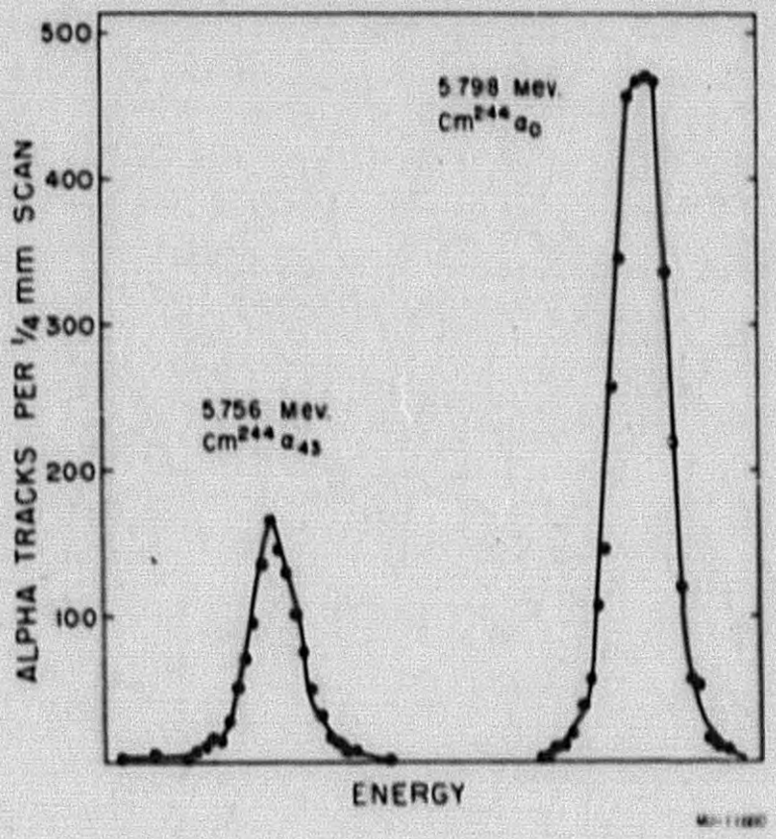

Fig. 9. High-resolution exposure showing main Cm^{244} alpha groups.

separation is obtained from the particle-energy separation by adding the energy due to the recoil correction (0.7 kev). The limit of error on the energy separation of ± 0.6 kev is a combination of two uncertainties. The uncertainty in locating the positions of the peaks on the photographic plate is taken as the average deviation of the values given in Table XI. This amounts to 0.2 kev. An additional 0.4 kev (1%) is added to account for the spectrograph dispersion calibration uncertainties. This is certainly a conservative limit of error. With 5.798 Mev taken as the energy of the main alpha group, the energy of the less intense group is 5.756 Mev.

From considerations of other even-even alpha emitters in this region, the highest-energy group (in all cases also the most abundant group) is considered the alpha transition from ground state to ground state.[6] Since the energy of this group in Cm^{244} decay is 5.798 Mev, the alpha-decay energy for this nuclide is 5.895 Mev. As the lower-energy group populates a level 42.9 kev above the level populated by the most intense transition, this second alpha group is denoted as α_{43}.

The abundance data on the two main groups is summarized in Table XII. In all cases the peaks were scanned completely (full scans taken at every 1/4 mm) and then integrated to determine their intensities. The ± 0.6% limit of error given in the table is the average deviation of the seven experiments. This is a somewhat larger limit than one gets by considering the standard statistical deviation for the seven runs including an estimated 3% alpha-track counting error. The abundances quoted in Table XII are contingent upon the absence of other fine-structure components of intensity of 0.1% or more intensity. The next section shows that this condition is satisfied.

This study of the main components of the Cm^{244} alpha spectrum has been somewhat more intensive than the earlier study by Asaro et al.,[9] and has led to revised intensities and a revised energy separation of the main groups. It should be pointed out that the new more exact values are certainly within the experimental uncertainties of the earlier results.

Table XI

Energy separation of the main Cm^{244} alpha groups

Exposure No.	Particle-energy separation (kev)	Decay-energy separation (kev)
345	42.5	43.2
347	41.8	42.5
348-II	42.3	43.0
348-III	42.2	42.9
349-II	42.5	43.2
349-III	42.0	42.7
352	42.0	42.7
Average value	42.2 ± 0.6 kev	42.9 ± 0.6 kev

Table XII

Intensities of the main alpha groups of Cm^{244}

Exposure No.	α_0 abundance (%)	$\alpha_{1,3}$ abundance (%)
345	77.3	22.7
347	77.3	22.7
348-II	76.0	24.0
348-III	76.8	23.2
349-II	75.9	24.1
349-III	77.4	22.6
352	76.1	23.9
Average abundances	76.7 ± 0.6%	23.3 ± 0.6%

2. New Alpha Group of Curium-244

As mentioned previously, one of the aims of the present study was to search for a low-intensity, lower-energy alpha transition common to many of the even-even alpha emitters in this region. Owing to the low geometry of the spectrograph, exposures of several days in length would be necessary to observe any low-intensity groups. All three sources were exposed for long periods of time in the course of this search. As expected, a group of small abundance was found about 100 kev lower in energy than α_{43}. Figure 10 indicates a spectrum obtained in this series of runs showing the low-intensity group. The data regarding this transition are summarized in Table XIII. There, for each exposure, the separation from α_{43} and the abundance of the new group are given.

Because of the very small intensity of this new group, the tailing effects of the various samples are rather important. Runs 343 and 346 were made with Sources A and B respectively, and the rest of the exposures were made with Source C. All the exposures were made with an 18-mil defining slit masking the source (high-resolution condition), with the exception of run 353, for which a 1/8-in.-wide slit system was employed. The exposure durations varied from 2 days for the higher-geometry run (353) to 7 days for run 488. It was expected from the preliminary experiments on the tailing effects that those exposures made with Source C would give the best peak-to-background ratio. As will become evident later, this was so.

Because of the tremendously large number of alpha tracks in the main peaks, they were not scanned completely in the vertical direction. Instead, a field of view or fraction of a field of view was counted in three different vertical positions for each horizontal position on the plate. The three counts were totaled and plotted for each horizontal position. It was felt that this method of partial scanning should give a pretty good determination of the peak position with a minimum of counting effort. However, judging from the data on the 1% alpha group in the Am^{243} spectrum, the tendency with this partial-scanning technique is to give a peak position displaced to a somewhat higher energy than the

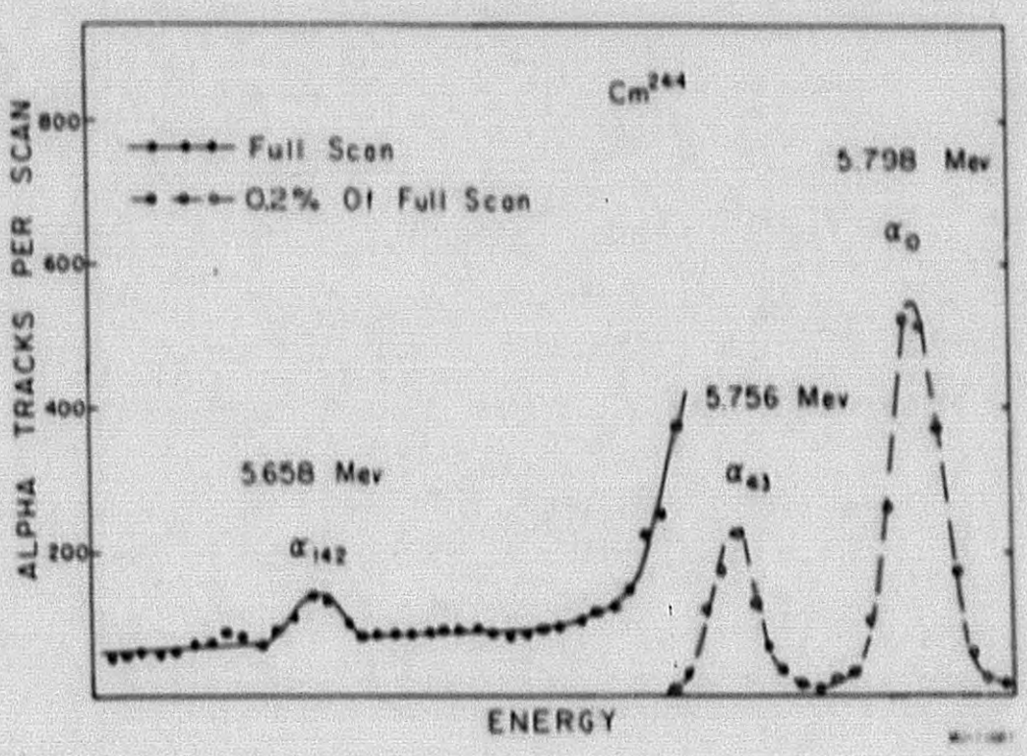

Fig. 10. High-resolution spectrum showing third alpha group of Cm^{244}.

actual peak position. This would lead to energy separations somewhat larger than the actual separations for Cm^{244}. The limited data available on this effect were not deemed sufficient to justify making an adjustment to the observed separations. The energy uncertainty is therefore increased somewhat.

After correction of the particle-energy separation given in Table XIII for the recoil energy, the decay-energy separation of the new group from α_{43} is 99.3 kev. Thus, the new group populates a level 142.2 kev above the ground state and will be called α_{142}. The energy of this new group is 5.658 Mev.

Table XIII

Low-intensity alpha group of Cm^{244}

Exposure No.	$\alpha_{142} - \alpha_{43}$ particle energy separation (kev)	α_{142} abundance (%)
343	96.9	1.1×10^{-2}
346	98.0	2.0×10^{-2}
351	98.4	1.5×10^{-2}
353	97.8	1.8×10^{-2}
488	97.2	2.0×10^{-2}
Best values	97.7 ± 1.5 kev	$(1.7 \pm 0.3) \times 10^{-2}$%

The large number of α_0 and α_{43} tracks somewhat complicates the abundance determination. To determine the number of α_0 and α_{43} tracks, short exposures (~10 min) were run at the same source setup immediately before or after the long exposures listed in Table XIII. The number of alpha tracks in the main groups on the long exposures were then calculated from the tracks recorded in the shorter runs. In arriving at the best value for the abundance of α_{142},

Experiments 351 and 488 were weighted doubly. This was considered justified from an analysis of the tailing effect. Those two exposures had by far the most favorable ratio of peak height to background. For example, in Exposure 343, a peak height of 50 tracks was observed on top of a 350-track background; but, in Exposure 351 this ratio was 60 to 200. The background in these cases is mostly due to tailing from the higher-energy groups of large abundance rather than to machine background. The uncertainty given in Table XIII is the average deviation from the best value.

The assignment of this new group to Cm^{244} decay should be considered somewhat. In the previous study by Asaro, a mixture of Cm^{242}, Cm^{243}, and Cm^{244} was run, and no alpha group was seen at 5.658 Mev (the energy of the new group). In these runs, the Cm^{243} and Cm^{244} activity were fairly comparable. The absence of the main Cm^{243} alpha groups from the current spectrum means that the new group at 5.658 Mev cannot be due to Cm^{243}. Cm^{242} is also eliminated as a possibility, since its activity is much less in the current samples than in those used in the previous study. Mass analysis of the current curium samples showed the presence of small amounts of Cm^{245} and Cm^{246} (of the order of 1% to 2% of each). Owing to the long half lives of these isotopes as compared with the half life of Cm^{244}, the intensities of any alpha groups belonging to the heavier curium isotopes would indeed be low. (Cm^{244} has a 19-year half life; Cm^{245} has a 1.15×10^4-year half life; and Cm^{246} has a 4.0×10^3-year half life[35, 36]). Cm^{245} has a main alpha group at 5.34 Mev.[37] Considering the reported Cm^{246} half life and alpha-decay systematics,[38] one would expect the main Cm^{246} alpha group (ground-state transition) to be at 5.37 Mev. This eliminates Cm^{246} as a possible assignment for the new group. A search in the region of these two energies using the current curium sample (Run 351) showed no alpha groups that could be attributed to Cm^{245} or Cm^{246}. Thus the possibility that the new alpha group is a high-energy transition belonging to Cm^{245} is eliminated on the basis of the absence of its main group. To check the possibility that the new alpha group might belong to short-lived components in the sample or daughters growing in, one of the exposures listed in Table XIII

(Run 488) was made 20 months after the others were made. One notes that the intensity of the new group relative to the main Cm^{244} alpha groups was constant over that period. These considerations make the assignment of the new alpha group at 5.658 Mev to Cm^{244} seem reasonable.

The alpha-particle data for Cm^{244} are summarized in Table XIV, where the absolute energy, decay energy to a_0, abundance, and partial alpha half life for each group are listed. The partial half lives are based on a value of 19 years[35] for the half life of Cm^{244}. The significance of these partial half lives with respect to alpha-decay theory is considered in a later section.

Table XIV

Complex alpha spectrum of Cm^{244}

Energy (Mev)	Decay energy to a_0 (kev)	Abundance (%)	Partial alpha half life (years)
5.798	0	76.7	24.8
5.756	42.9	23.3	81.5
5.658	142.2	0.017	1.1×10^5

3. Gamma Rays in Decay of Curium-244

A study was conducted of the main features of the gamma spectrum associated with the alpha decay of Cm^{244}, in the hope that information along this line would lead to the assignment of spins and parities to the levels involved. Since the only samples of Cm^{244} available also contained appreciable amounts of other curium isotopes, it was not possible to observe the Cm^{244} gamma spectrum directly. The approach used in this study was to determine the spectrum of the mixed isotopes and then subtract out the contributions from isotopes other than Cm^{244}. Two samples of quite different isotopic content

were used. One Cm^{244} sample was a portion of one of the spectrograph sources used for the measurements just discussed. The other sample contained a much larger percentage of Cm^{244}.

The spectrum obtained on the first sample of mixed curium isotopes with a NaI detector coupled to the 50-channel analyzer is shown in Fig. 11. This sample was shown to contain 73% Cm^{244} and 27% Cm^{242} activitywise by a pulse-height analysis using an ion chamber. Considering the method of preparation of the sample, it was expected that small amounts of Cm^{243} would be present and that the gamma rays from the decay of Cm^{243} would be seen in the spectrum. Considering the similarity in the alpha spectra of Cm^{242} and Cm^{244}, one would expect these two isotopes to have very similar gamma radiations. Considering the data discussed previously for Cm^{242}, the Cm^{242} and Cm^{244} in this sample should contribute to peaks at ~ 44, ~ 100, and ~ 150 kev. The known gamma rays of Cm^{243} would be seen at 104, 226, and 278 kev.[9] Thus, the 43-kev radiation seen in Fig. 11 is probably due to Cm^{242} and Cm^{244}. All three isotopes contribute to the 100-kev peak, and it will be evident later that this peak is mostly Cm^{243} radiation. The small peak at ~ 150 kev is also expected to come from both Cm^{242} and Cm^{244}. The radiation at 65 kev is probably due to platinum K x-rays produced by the fluorescence excitation of the platinum backing. Further out in the spectrum, and not shown in Fig. 11, are the two Cm^{243} gamma rays at 226 and 278 kev.

The Cm^{243} contribution to this spectrum was subtracted out by running a sample of Cm^{243} and normalizing the two high-energy gamma rays between the two runs. The result of this subtraction is shown in Fig. 12. (Note that the 65-kev region has been arbitrarily omitted in this spectrum.) Next, the Cm^{242} contribution to this new spectrum was determined. An accurately alpha-counted Cm^{242} sample was run at the same geometry as the mixed curium sample. As the exact alpha-counting rate of the sample of mixed curium isotopes and the percentage (activitywise) of Cm^{242} in this sample were known, the Cm^{242} contribution to Fig. 12 could be easily determined. The result of this subtraction should represent the Cm^{244}

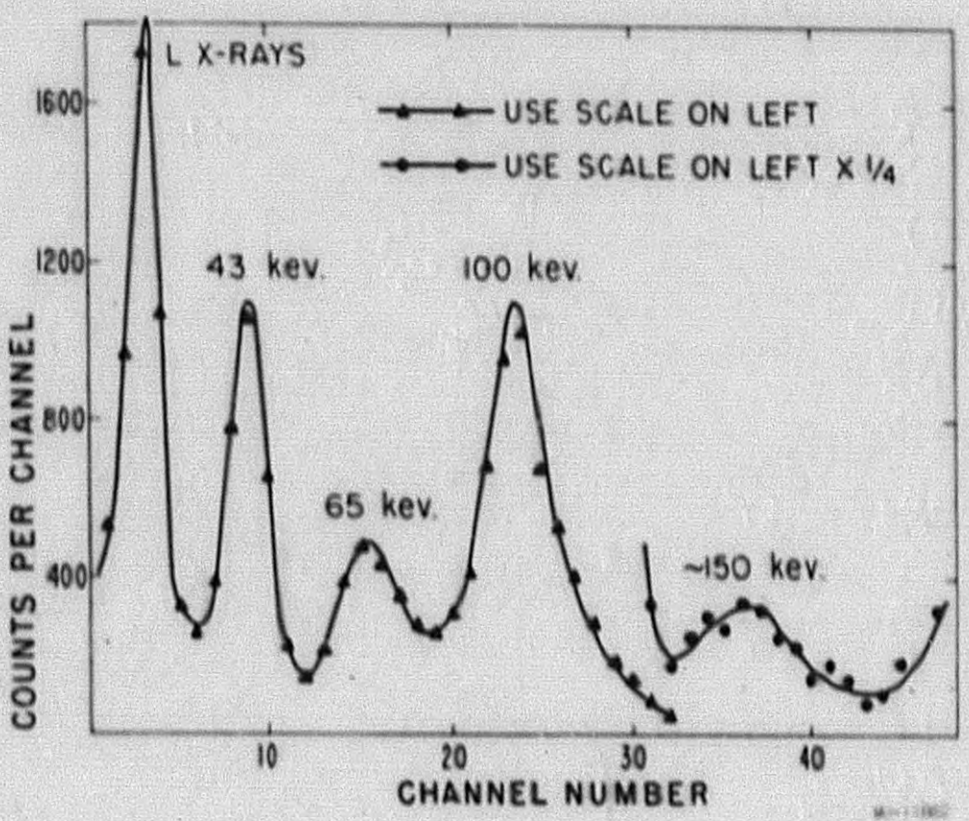

Fig. 11. Gamma-ray spectrum of sample of mixed curium isotopes (low Cm^{244} content).

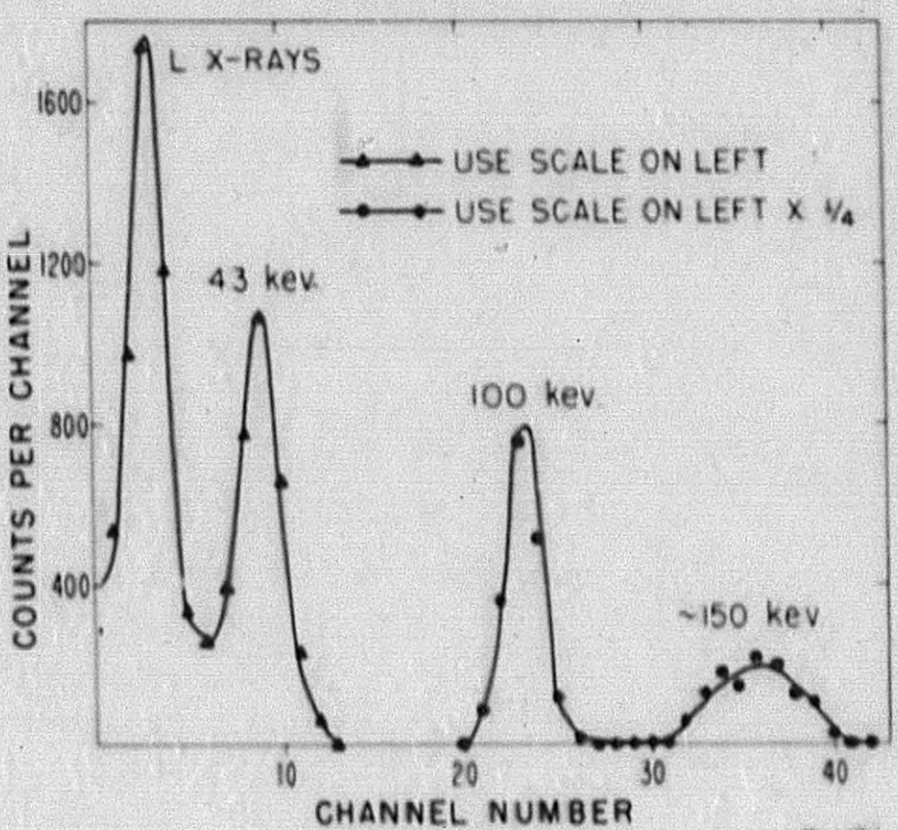

Fig. 12. Gamma-ray spectrum of sample of mixed curium isotopes after removal of Cm^{244} contribution.

gamma spectrum. As no accurate geometry determination was made during the course of this experiment, the abundances of the Cm^{244} gamma rays were first measured with respect to the abundance of the 44-kev gamma ray in Cm^{242} decay. (The necessary corrections to the raw data were made as indicated in the section on experimental techniques.) Using the new Cm^{242} abundance data (previous section), the abundances of the gamma rays in Cm^{244} decay were then calculated and found to be 2.1×10^{-4} (43-kev gamma ray), 1.5×10^{-5} (100-kev gamma ray), and 1.3×10^{-5} (150-kev gamma ray). A second determination of the most prominent 43-kev gamma ray gave an abundance of 2.0×10^{-4} photons per alpha particle.

The second sample used in this study contained 98.2% Cm^{244} and 1.8% Cm^{242} (activitywise by alpha-particle pulse analysis). The Cm^{243} content in this sample was smaller than in the previous sample. As this sample was made by a longer neutron irradiation than the previous sample, Cm^{245} and Cm^{246} should be present to appreciable extents. Mass analysis of the curium confirmed this, showing ~4% Cm^{246} and ~2% Cm^{245} present (masswise). Since Cm^{246} is an even-even isotope and has a much longer half life than Cm^{244}, the Cm^{246} radiations would not be expected to contribute much to the gamma-ray spectrum. On the other hand, one might expect that Cm^{245} gamma rays would contribute to a small extent to the observed spectrum.[39] Figure 13 shows the gamma spectrum of this sample taken in the same way as in the previous case. Again, the 226- and 278-kev gamma rays of Cm^{243} are off scale.

The Cm^{243} and Cm^{242} contributions were determined and subtracted out in the same way as in the previous case. The resulting spectrum is shown in Fig. 14. One sees the expected 43-, 100-, and 150-kev gamma rays of Cm^{244}. Also visible are high-energy tails on the 100- and 150-kev peaks. These are interpreted as being due to the Cm^{245} in the sample. Asaro[39] has examined the Cm^{245} gamma-ray spectrum, and he reports K x-rays and 130- and 172-kev gamma rays as the prominent features. In the determination of the abundances of the Cm^{244} gamma rays, the peaks were integrated with the high-energy tails excluded. Also, the Cm^{245} K x-ray contribution to the

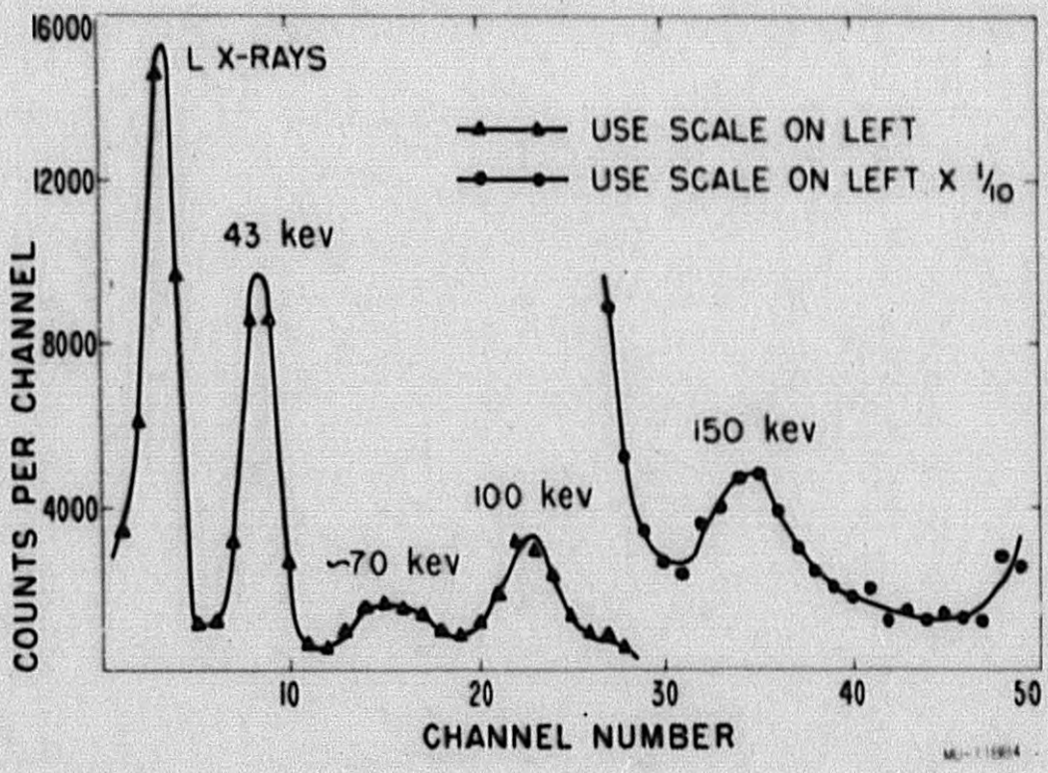

Fig. 13. Gamma-ray spectrum of sample of mixed curium isotopes (high Cm^{244} content).

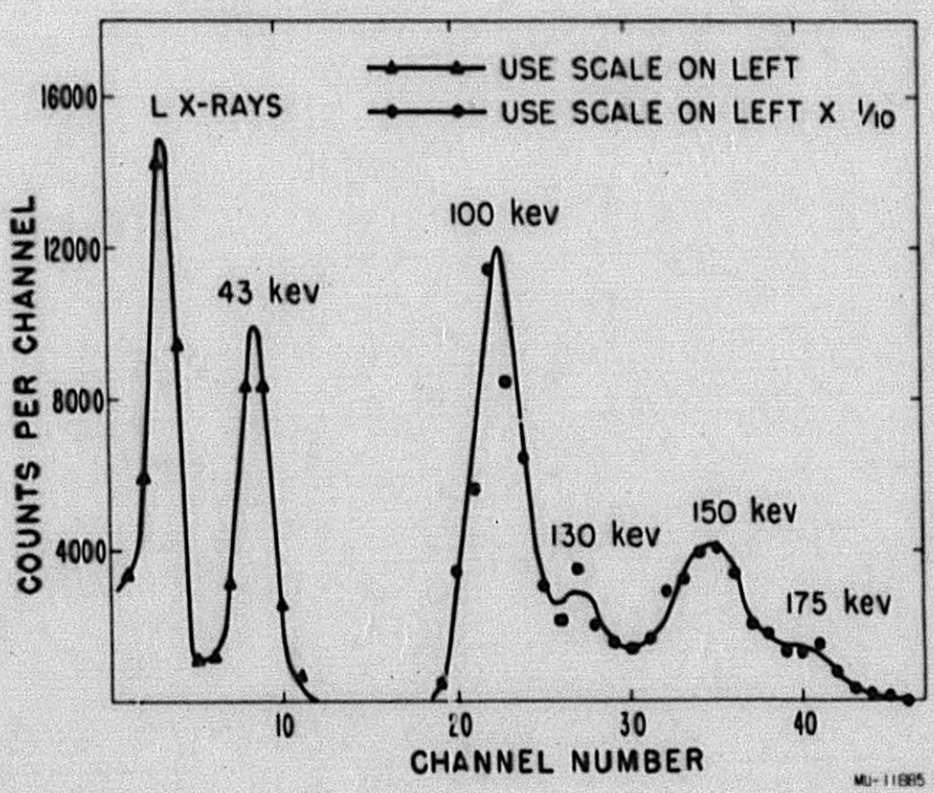

Fig. 14. Gamma-ray spectrum of sample of mixed curium isotopes after removal of Cm^{242} and Cm^{243} contributions.

100-kev peak was estimated from the available decay data and subtracted from the 100-kev peak. This estimation was made by resolving out a 172-kev gamma ray and integrating its total counts. This number was multiplied by six to get the number of K x-rays. (Asaro[39] reports 14% 172-kev radiation and 68% to 93% K x-rays in Cm^{245} decay).

The absolute geometry of the counting arrangement was determined by running an accurately counted Am^{241} sample. The known abundance of the prominent 60-kev gamma ray (0.4 gamma rays per alpha particle)[23] was used here as the standard. Thus, absolute intensities of the Cm^{244} gamma rays were determined directly. They are 2.0×10^{-4} (43-kev gamma ray), 1.3×10^{-5} (100-kev gamma ray), and 1.4×10^{-5} (160-kev gamma ray). The agreement between these values and the ones obtained with the first sample is good. It should be pointed out that the intensity of the 100-kev gamma ray is probably the poorest of the three, considering the large corrections made in this region due to the Cm^{243} and Cm^{245} in the samples.

4. Decay Scheme of Curium-244

The decay scheme suggested by these data is given in Fig. 15. The first two excited states are quite well defined by the alpha-spectrum work. The 43-kev gamma ray most certainly de-excites the first excited state. The 100-kev gamma fits well as the transition between the first and second excited states. (The placement of the 150-kev gamma ray is discussed presently.)

Using the alpha-particle abundance to the first excited state (23.3%) and the intensity of the 43-kev gamma ray (2.0×10^{-2}%), one calculates a conversion coefficient of 1160 for the 43-kev gamma ray. This represents the total L, M, N, etc. conversion. Smith and Hollander[34] have observed conversion lines of a 42.88-kev gamma ray in Cm^{244} decay, and report an L/M/N ratio of 2.5/1.0/0.3. Using this ratio, the L-conversion coefficient is 760. This value is in the expected range of an E2 transition on the basis of extrapolations from the data of Gellman, Griffith, and Stanley.[40] Also, Smith and Hollander have made an E2 assignment for this gamma ray on the basis of the

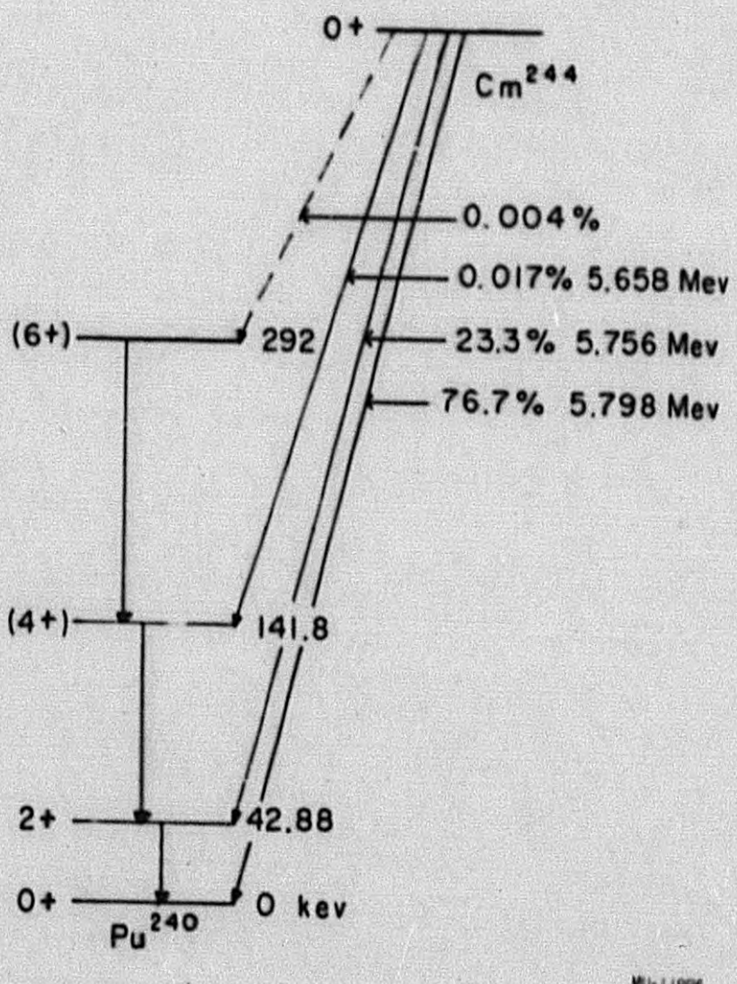

Fig. 15. Decay scheme of Cm^{244}.

observed relative L-subshell ratios. Since the ground state of the even-even Pu^{240} is undoubtedly 0+, the first excited state is 2+.

The 100-kev photon de-exciting the level populated by α_{142} is in 1.4×10^{-3}% abundance. Since the alpha population to that level is 1.7×10^{-2}% the conversion coefficient for the 100-kev gamma ray is 11. On this basis alone, it is rather difficult to choose between an M1 or E2 assignment using the predicted L-subshell conversion coefficients from Gellman et al.[40] However, the same gamma ray has been observed in the study of the conversion electrons from Am^{240}, in which case the relative L-subshell ratio clearly indicates an E2 assignment.[41] The energy of this transition, from the electron study, is 98.90 kev. The energies from the conversion-electron studies cited here are considered more accurate than either the alpha-particle or gamma-ray data; therefore they have been used in Fig. 15. A 4+ assignment for the second excited state is preferred, although one cannot rule out other assignments.

The assignment of the 150-kev gamma ray is made solely on the basis of the collective model. It places the next level at about the right energy, as expected for the 6+ state in well-developed rotational band.[42] This assignment would make the 150-kev gamma ray an E2, and the expected conversion coefficient would be about 2. From the intensity of this gamma ray (1.4×10^{-5}), the direct alpha population to the 6+ level would be $\sim 4 \times 10^{-5}$ (about 1/4 the abundance of α_{142}). This intensity would be just on the verge of detection in the experiments just described.

A well-developed rotational band is apparent in the decay of Cm^{244}. Some of the features of this band, especially the degree of alpha population to the various members, are discussed in a later section.

D. Alpha Decay of Protactinium-231

One of the natural alpha emitters that has been the subject of a considerable amount of study is the long-lived Pa^{231}. Pa^{231} has a half life of 3.43×10^4 years,[43] and decays into the predominantly beta-decaying 22-year Ac^{227}. Thus, one can work with the Pa^{231} for fairly long periods of time with little trouble from the daughter activities.

The only previous alpha-particle work on this nuclide was done by the French group (Rosenblum),[44] which showed six alpha groups attributable to Pa^{231}. A group of Russian workers carrying out alpha-particle spectroscopy on Pa^{231} at the same time as this work have published some preliminary results, which show that the alpha spectrum is considerably more complex than at first thought.[45]

There has been a considerable amount of work done on the gamma radiations and the conversion electrons accompanying this decay. Scharff-Goldhaber and MacKeown[46] studied the low-energy gamma rays by the use of proportional counters. Riou[47] used absorption techniques with proportional and Geiger counters to study the gamma spectrum up to 300 kev. Meitner[48] did the first conversion-electron study in 1928. Teillac[49] used a cloud chamber to study the prominent conversion electrons. The most recent and complete study of the conversion-electron spectrum was done by Falk-Vairant.[50] And, a thorough gamma-ray study including many coincidence experiments was concluded by Moore[51] in 1953. As these studies progressed, the extreme complexity of this decay became more and more evident. Many of the above-mentioned investigators suggested decay schemes for Pa^{231}. As becomes more evident in the next few pages, the absence of extremely good-resolution alpha-particle spectroscopy made the interpretation of the data somewhat difficult and venturesome, so that the suggested decay schemes have not held up.

The Pa^{231} used in this study was made by a long neutron irradiation of Th^{230}. This produces predominantly the 25-hour beta-emitting Th^{231}, which decays to give Pa^{231}. Owing to the long half life of Pa^{231}, an extremely long irradiation is necessary to build up much Pa^{231} activity.

From consideration of the half lives of the other protactinium isotopes produced in this type of bombardment, Pa^{231} is easily gotten in a state of high radioactive purity by allowing the protactinium to stand for several months. When this study began, the Pa^{231} had stood about two years since purification, so that there was a considerable amount of alpha activity due to the alpha-emitting daughters of Pa^{231}. The sample was used as such for a considerable amount of the alpha-particle spectroscopy. Later, for gamma-ray studies and confirmatory alpha-particle studies, a sample of the old protactinium was repurified, removing the daughter activity.

The chemistry used in the protactinium purification consisted of running the mixture through a column containing Dowex A-1 anion-exchange resin in an 8 M HCl solution and washing for several column volumes. The protactinium was then eluted off with 3.6 M HCl. This procedure was varied later by eluting the protactinium with 0.1 M HI in 7 M HCl. This procedure gave protactinium fractions of 99+% purity.

1. Alpha Spectrum of Protactinium-231

Three spectrograph samples were made by vacuum sublimation of the protactinium, which contained about 5% of each daughter. One sample containing 4×10^5 alpha disintegrations per minute (4 micrograms of Pa^{231}) was extremely thin. The other two contained larger amounts of activity and mass, having count rates of 1.8×10^6 and 3.5×10^6 alpha disintegrations per minute. Later, another vaporized sample of 8×10^5 alpha disintegrations per minute was prepared from the purified Pa^{231}. The majority of the experiments on this isotope were done with the first three samples, the fourth being used primarily for confirmatory purposes. Several different geometry arrangements were used in the course of the many runs, and these are discussed for each experiment.

The first two exposures were made with the 4-μg sample, and were made mostly as exploratory runs. They showed the main features of the spectrum reported by Rosenblum (all groups in 10% abundance or larger appearing). Rosenblum's spectrum is summarized in

Table XV, and it is used as an orientation guide in discussing the new experiments. Although the resolution on the first runs was poor (maximum-geometry slit arrangement), there was distinct evidence that the group reported by Rosenblum at 5.002 Mev (47%) was complex and probably composed of two groups about 18 kev apart (see Fig. 16). The higher-energy component of this doublet appeared to be present in a slightly smaller abundance than the lower-energy group. The possibility that some low-intensity groups also exist in this region is no yet ruled out.

Table XV

Pa^{231} complex alpha spectrum (Rosenblum)

Energy (Mev)	Abundance (%)
5.042	11
5.002	47
4.938	25
4.838	3
4.720	11
4.660	1-3

This same sample was exposed at high resolution (18-mil defining slit) for 4 days to further examine the complex peak. It would be expected that only the most prominent features of the spectrum would appear on this run, owing to the small amount of activity on the plate; this was the case, as only the groups in 20% or larger abundance had peak heights larger than the machine background. The peak at 5.002 Mev, suspected of being complex, was resolved completely on this run into two alpha groups separated by 15.8 kev and in roughly equal abundances (see Fig. 16). The previously reported 25% group at 4.938 Mev also appeared in abundance comparable to each component of

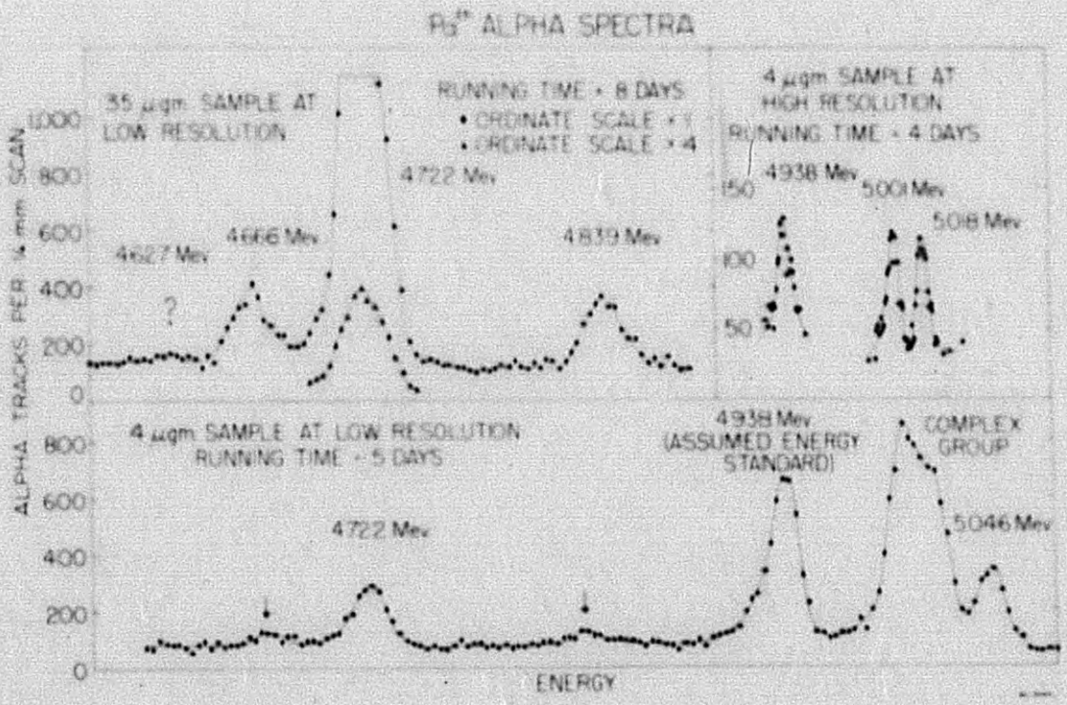

Fig. 16. Main features of the Pa^{231} alpha-particle spectrum.

the double group. In the absence of an energy standard on these runs, the group at 4.938 Mev (Rosenblum) was used as a standard for an early publication of this work.[52] As is shown later, this energy has been confirmed. From this high-resolution run, the members of the doublet would be at 5.018 and 5.001 Mev. The highest-energy group in the Pa^{231} spectrum is at 5.042 Mev (Rosenblum). This group also appeared on this run, but owing to its low peak height with respect to background, the possibility that this group is complex could not be eliminated. This matter is discussed more fully in connection with later experiments.

This same sample was run with a Ra^{226} sample at maximum geometry to check the energy determination. The Ra^{226} group at 4.777 Mev and the Po^{210} group at 5.299 Mev were used as energy standards.[6] Since the two samples were of widely different masses, there was a large difference in the half widths of the peaks involved here (almost 10 kev). After correction for this difference, the energy of the group previously used as an energy standard was 4.939 Mev, in excellent agreement with Rosenblum's value of 4.938 Mev. The value of 4.938 Mev is used here, as was done in the experiments already discussed.

To examine the lower-intensity components of the Pa^{231} spectrum, the largest sample was employed. This source (3.5×10^6 alpha disintegrations per minute) was run for 9 days at the maximum-geometry slit arrangement (see Fig. 16). The groups previously reported as being in 3% (4.838 Mev) and in 1% to 3% (4.660 Mev) abundances were confirmed, but their intensities were definitely less than 3%. Also, the possibility of a new group's existing at 4.627 Mev in about 0.3% abundance appeared on the long exposure. Since the peak height of this group was small compared with the background in this region (35 tracks versus 125 tracks background), the presence of this group is only tentative. A limit of 0.3% can be set on the abundance of other groups down to 4.41 Mev.

A 12-day rerun of the most intense sample at maximum geometry was undertaken to check the tentative low-intensity group. The peak appeared in the same intensity again, but again background considerations make it difficult to conclude that it is an actual alpha group. The

possibility exists that this peak can be due to some kind of sample effect. Until Pa^{231} samples are run in the double-focusing alpha-particle spectrograph, the assignment of this group will have to be considered tentative.

Up to this point, the alpha-particle studies presented here have essentially confirmed the spectrum reported by Rosenblum, with three exceptions: (a) the 47% group reported by Rosenblum is actually two groups, (b) the low-intensity groups reported by Rosenblum are in lower abundance than previously thought, and (c) there is a possibility of a new low-intensity low-energy group belonging to the spectrum.

Next, a 5-day exposure at maximum geometry with the 18-μg sample showed additional complexity to the spectrum. Indications of three new groups were present. These groups are either between fairly intense groups or in the low-energy tail of an intense group, so that they would not have been observed in the previous runs. The runs with the small sample would not have shown them because of insufficient activity. The tailing in the large sample obscured them on the runs with that source. To check the presence of these new groups, this same sample was rerun with the same slit arrangement, producing the same spectrum. Figure 17 shows this spectrum, with arrows indicating the locations of the new groups.

The same sample was then exposed with the 18-mil defining slit for 6 days in the hope that this would allow the resolution of the most intense of these new groups. This completely resolved a group of 2% abundance between the low-energy group of the doublet and the group at 4.938 Mev (Fig. 18). This group would be about 75 kev lower in energy than the highest-energy group (presumably the ground-state transition). Also, a group of about 3% abundance was partially resolved from the low-energy tail of the 4.938-Mev group. The other new group was of too small intensity to be seen on this run. One also notes in Fig. 18 that the doublet is almost completely resolved. Furthermore, the highest-energy group in the spectrum is completely resolved and appears to be a single group.

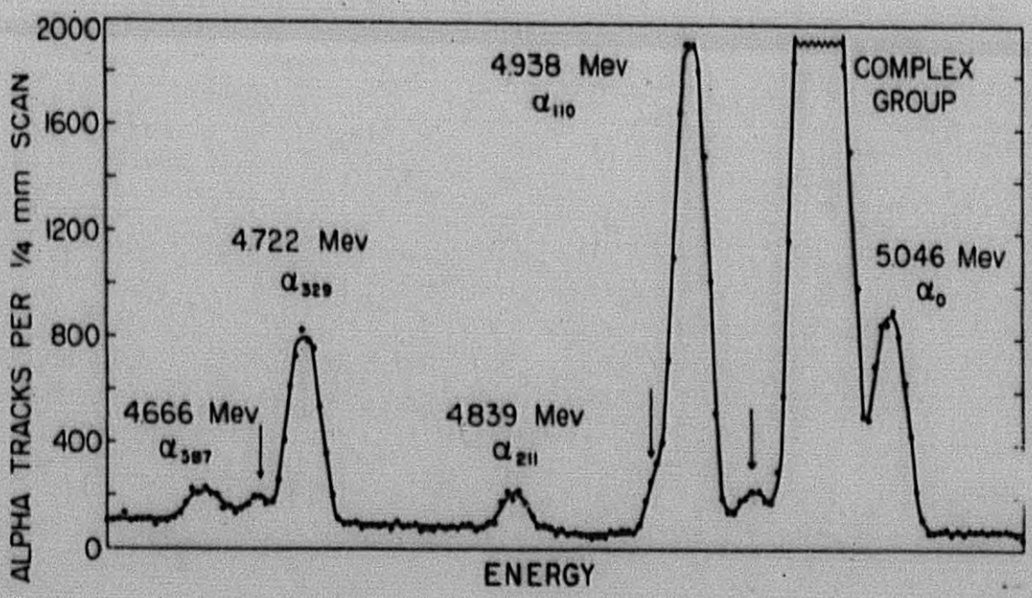

Fig. 17. Alpha-particle spectrum of Pa^{231} at low-resolution conditions.

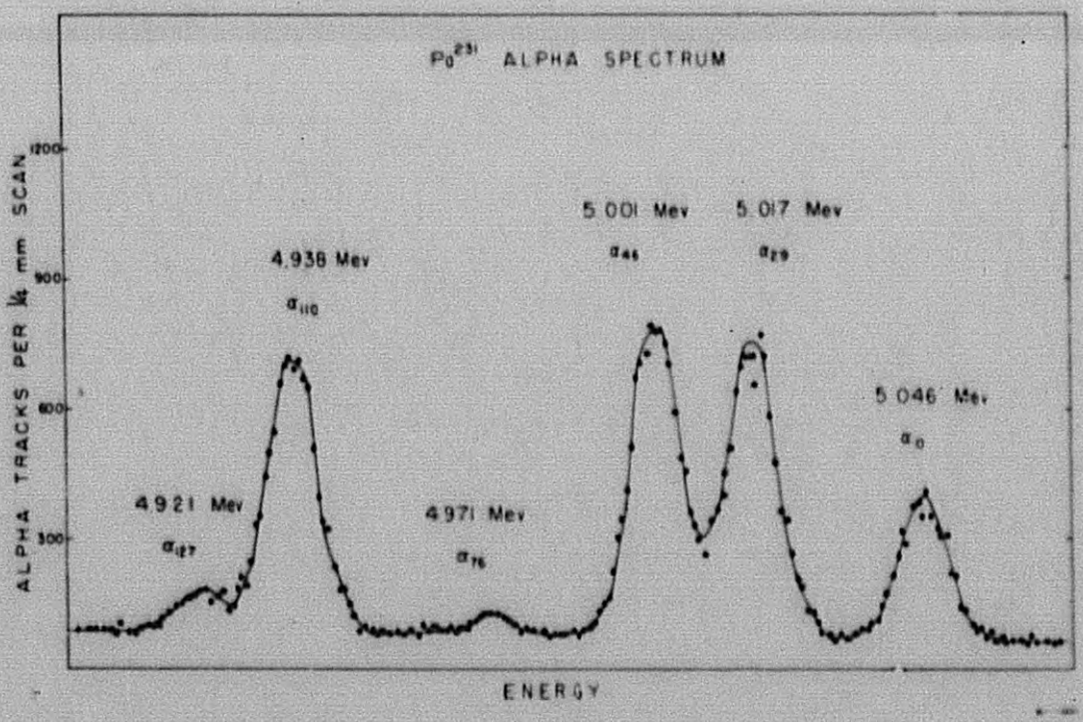

Fig. 18. Partial Pa^{231} alpha-particle spectrum at high resolution.

This same sample was then exposed for 5 days with a 1/16-inch slit arrangement. It was hoped that this would allow better resolution with only a small loss in peak height compared with the maximum-geometry run discussed previously. This was necessary in order to confirm the third low-intensity group indicated in Fig. 17. The desired effects were obtained, and excellent confirmation of the new groups resulted (Fig. 19). The arrows in Fig. 19 indicate the locations of the new groups.

Consideration of the activities present in the sample leads one to believe that these new groups definitely belong to Pa^{231}. This is so, as none of the daughter alpha emitters have known alpha groups in the energy regions of the new groups. To confirm the assignment of these alpha groups to Pa^{231}, the source of the purified protactinium was exposed in the spectrograph. Two exposures were made, one with the 18-mil slit and the other with the 1/16-inch slit. The new groups suspected of belonging to Pa^{231} were all present in the expected intensities. Thus, the assignment of the three new alpha groups to Pa^{231} seems quite certain.

The data from these spectrograph runs are summarized in Tables XVI through XVIII. Table XVI gives the energy separations observed between various alpha groups. Whenever possible, the separation from the 4.938-Mev group was measured. In some of the exposures, the high-energy portion of the spectrum was not observed, and the separations from the most abundant of the low-energy groups were determined. The best values listed in Table XVI were arrived at through consideration of the conditions under which each exposure was made, i.e., sample size, duration of exposure, and the resolution obtained. The 0.3% group is not listed in the table, because of its highly tentative nature. Also, there are listed several miscellaneous separations that are of use later when the decay scheme is discussed.

The abundance data are given in Table XVII. Information is listed only for those cases in which groups were clearly discernible and sufficiently resolved. The abundances are listed relative to the α_{110} plus α_{127} abundance, since in most cases those two groups were not

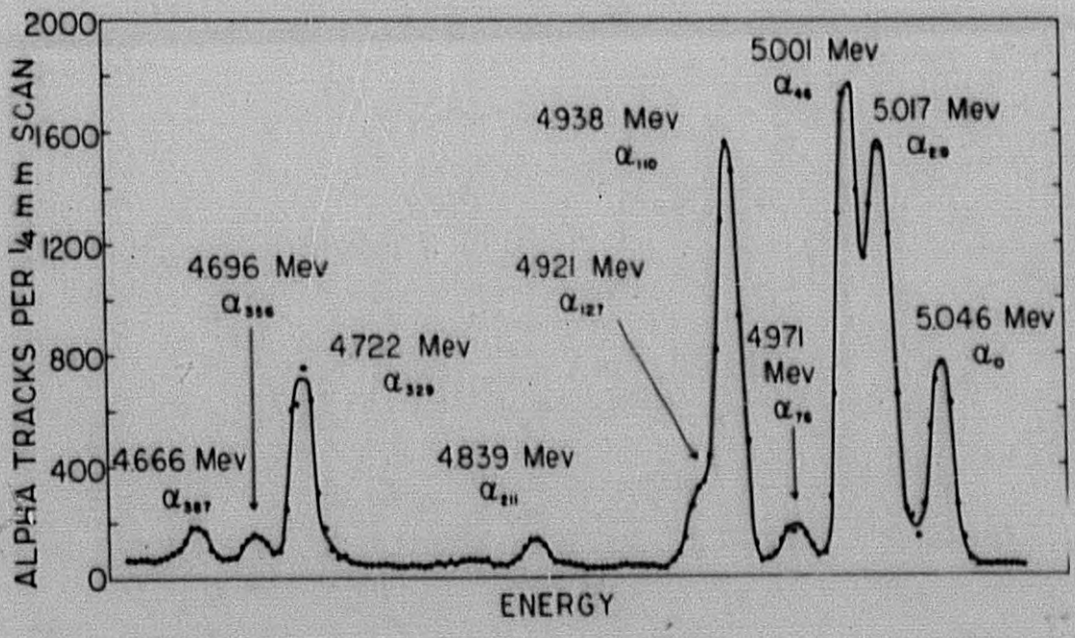

Fig. 19. Pa^{231} alpha-particle spectrum at medium resolution.

resolved. In some cases, where near-by groups could not be completely resolved, they were integrated together, and the abundance compared to α_{110} plus α_{127}. Although data taken in this way are not included in Table XVII, they were considered in arriving at the best values. Since there are rather large variations noted in some of the intensity ratios, it is felt that abundances having more than two significant figures have no meaning.

Table XVIII summarizes the complex alpha spectrum of Pa^{231}. There the absolute energy, decay-energy separation from the highest-energy group (the assumed α_0), percentage abundance, and partial alpha half life are given for each transition. These data are easily compared with those from Rosenblum (Table XV). As mentioned previously, Russian workers[45] have studied the complex alpha spectrum of Pa^{231}. They report eight alpha groups and indicate a tentative assignment of a ninth. The ninth group corresponds to α_{356}, which has been completely resolved in this study. The Russian workers have not reported a group corresponding to α_{127}. This would have been obscured in their work because of tailing from the α_{110} peak. Also, they have not been able to completely resolve the double peak at about 5.01 Mev as yet.

2. Gamma-Ray Studies on Protactinium-231

A study of the principal gamma rays accompanying the decay of Pa^{231} was also undertaken. As mentioned earlier, there has been a considerable amount of research along this line in the past. As the various experiments are discussed, a comparison with the existing data is given.

For the gamma-ray studies that follow, the stock protactinium containing Pa^{231} and a considerable amount of daughter activity was purified by the procedure already discussed. The samples prepared in this way were examined immediately for gamma radiation, so that little chance for interference from the daughter radiations exists.

Table XVI

Energy separations of Pa^{231} alpha groups

Exposure	Separations from the 4.938-Mev group (a_{110}) (in kev)									Miscellaneous separations			
	a_0	a_{29}	a_{46}	a_{76}	a_{127}	a_{211}	a_{329}	a_{356}	a_{387}	$a_{356}-a_{329}$	$a_{387}-a_{329}$	$a_{211}-a_{329}$	a_0-a_{29}
321	109.1	81.0	63.5				214.1						28.0
324						98.2	215.9		273.1		57.2	117.7	
325		79.8	64.0										
392											56.2	118.2	
406	109.4			32.2		99.5	216.8	244.2	274.3	27.4	57.5	117.3	
435	107.4	78.5	62.2	34.2	16.2								28.9
436	107.8			32.5		99.2	214.0	240.6	269.1	26.5	55.1	114.8	
441	108.3			32.3	18.1	98.0	215.0	239.2	269.6	24.2	54.6	117.0	
490	108.2	79.1	62.2	33.7	17.9	100.6	216.3	242.1	273.9	25.8	57.6	115.8	29.1
Best values (kev)	108.2	79.1	62.9	33.2	16.9	98.9	215.4	241.4	271.8	26.0	56.4	116.5	28.9

Table XVII

Pa^{231} alpha group abundance data

Exposure	Abundance relative to $a_{110} + a_{127}$									
	a_0	a_{29}	a_{46}	a_{76}	a_{110}	a_{127}	a_{211}	a_{329}	a_{356}	a_{387}
324							0.055			
406							0.052	"		
435	0.43	0.90	0.96		0.89	0.11				
436							0.052			
441	0.40			0.091			0.052	0.42	0.055	0.083
490	0.44			0.092			0.063	0.44		
Best values	0.42	0.90	0.96	0.091	0.89	0.11	0.055	0.43	0.055	0.083

Table XVIII

Complex alpha spectrum of Pa^{231}

Energy (Mev)	Decay energy to a_0 (kev)	Abundance (%)	Partial alpha half life (years)
5.046	0	10	3.4×10^5
5.017	29.4	23	1.5×10^5
5.001	46.1	24	1.4×10^5
4.971	76.3	2.3	1.5×10^6
4.938	110.1	22	1.6×10^5
4.921	127	2.8	1.2×10^6
4.839	211	1.4	2.5×10^6
4.722	329	11	3.1×10^5
4.696	356	1.4	2.5×10^6
4.666	387	2.1	1.6×10^6

The low-energy portion of the gamma-ray spectrum was examined with a proportional counter. Alpha particle–gamma ray coincidences were run with a ZnS screen serving as the alpha detector. The spectrum up to 40 kev was observed, showing the various L x-ray peaks and a very prominent 27.5-kev gamma ray. As a very careful calibration of the energy scale was made, with an Am^{241} source, the energy uncertainty in this measurement is ± 0.5 kev at the worst. This energy measurement is in good agreement with those by Scharff-Goldhaber,[46] Riou,[47] and Moore.[51] All these have reported a prominent 27-kev gamma ray. Riou has reported the abundance of the 27-kev gamma ray as 9%.

Next, the gamma-ray spectrum up to 400 kev was studied with a NaI detector. The gamma-ray spectrum in coincidence with the alpha particles of Pa^{231} is shown in Fig. 20. Present in the spectrum are radiations of 300, 145, 95, and 65 kev. The 65-kev radiation is probably mostly due to platinum K x-rays, as this sample was mounted on platinum. The 95-kev radiation is probably actinium K x-rays. However, the presence of gamma rays in these regions due to Pa^{231} decay cannot be eliminated. The 145-kev peak is interpreted as due to Compton scatter of the 300-kev gamma ray. With Hg^{203} as a standard, (279-kev gamma ray),[43] it was shown that the energy of the prominent gamma ray was 300 kev, and not 280 kev as reported by Moore.[51] Riou has also reported a 300-kev gamma ray. Also, on the basis of the Hg^{203} gamma-ray half width, the 300-kev peak is definitely complex. Three gamma rays have been resolved out of the 300-kev peak, which have energies of ~ 265, 300, and ~ 330 kev. The resolution of the gamma rays is shown in Fig. 20. The abundance of the 300-kev gamma ray was 0.05 per alpha particle, and the abundances of the ~ 265- and ~ 330-kev gamma rays were about 1% for each. Riou reports a 4% abundance for the 300-kev gamma ray.

Coincidences were run between the 27-kev gamma ray and the rest of the gamma spectrum, with particular emphasis placed on the 300-kev region. The coincidence spectrum is shown in Fig. 21. Peaks were observed at 65, 145, and 300 kev. The 65-kev radiation is again

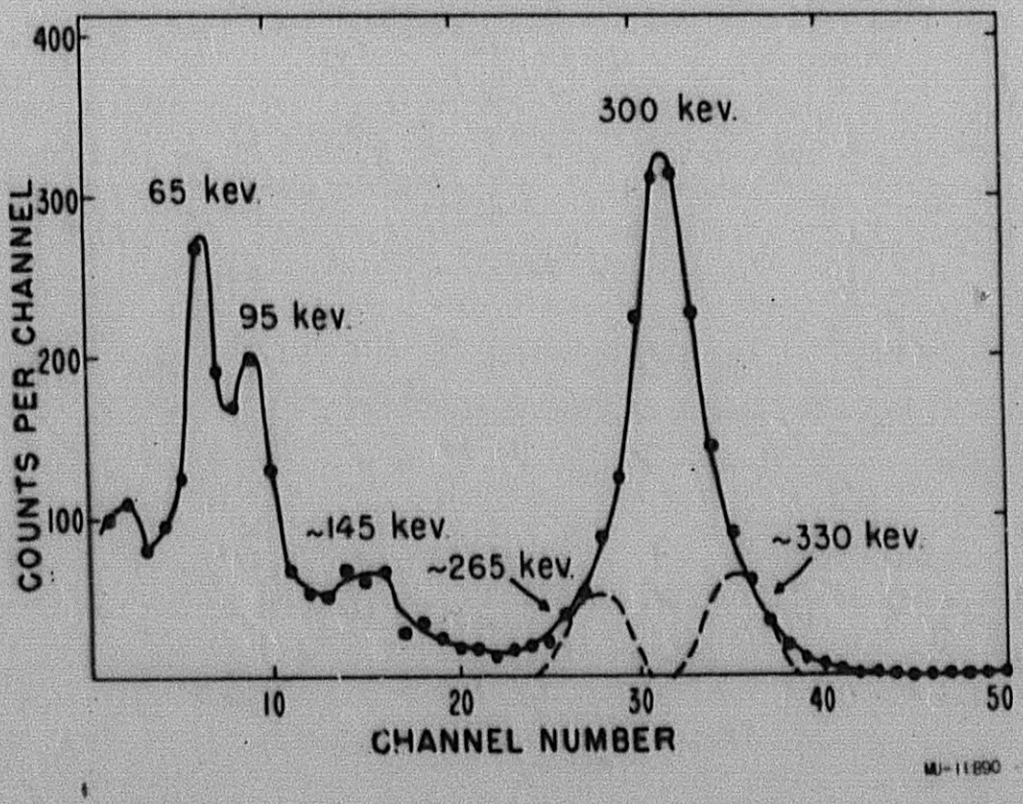

Fig. 20. Pa^{231} alpha particle-gamma ray coincidence spectrum.

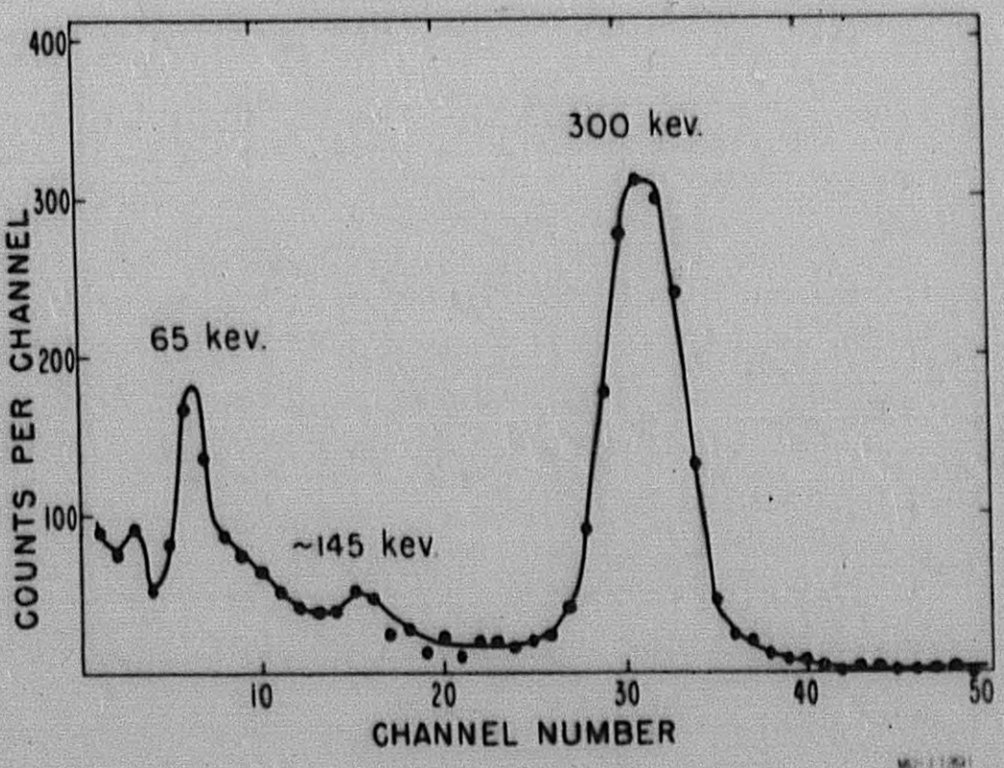

Fig. 21. Pa^{231} gamma-ray spectrum in coincidence with the 27-kev gamma ray.

probably due to platinum K x-rays, and the 145-kev peak is probably due to Compton scatter of the 300-kev gamma ray. One notes that the actinium K x-rays are not nearly as abundant as in the α-γ coincidence spectrum in Fig. 20. Also, the high-energy portion of the 300-kev peak (~330-kev gamma ray) does not appear to be in coincidence with the 27-kev gamma ray.

Coincidences were also run between the 300-kev peak and the gamma spectrum up to 130 kev (Fig. 22). As expected, the 27-kev gamma ray appeared in coincidence. (The L x-rays were absorbed out with aluminum and did not register). Also, the K x-rays were not observed, and a limit of 6×10^{-5} K x-rays per 300-kev gamma ray was set on their maximum coincidence intensity. This is in direct opposition to Moore's results,[51] which showed a K x-ray-280-kev gamma ray coincidence intensity of $\sim 1.5 \times 10^{-2}$.

Moore has done a more complete coincidence study of the gamma rays in this decay, including delayed-coincidence experiments. The results of his study are considered in more detail, in connection with the discussion of the decay scheme, in the next section.

Another type of coincidence study made in the course of this work was done with the aid of the new double-focusing alpha-particle spectrograph. The spectrograph source giving 1.8×10^{6} alpha disintegrations per minute and containing Pa^{231} and its daughters (vaporized onto aluminum) was used as a source in this case. Prior to the coincidence studies, the alpha spectrum from this source was obtained by varying the magnetic field and making counts of the alpha particles striking the ZnS detector as a function of the field strength. The spectrum obtained in this way had a resolution comparable to the high-geometry runs with this sample on the low-geometry spectrograph. The experimental setup for using the spectrograph in coincidence studies has already been discussed in an earlier section. One procedural difficulty was encountered in this experiment that makes the results somewhat hard to interpret. This difficulty was a depression of the energy scale on the 50-channel analyzer, presumably caused by the extremely intense gamma radiation striking the NaI crystal. Because of this depression, a good calibration of the energy scale could not be obtained.

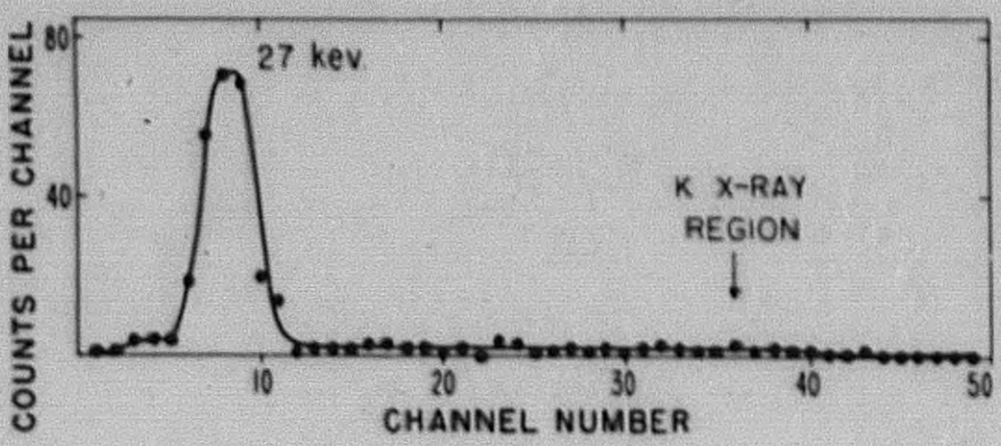

Fig. 22. Pa^{231} gamma-ray spectrum in coincidence with the 300-k radiation.

Coincidences were first run between the complex peak of α_{29} and α_{46} and the gamma-ray spectrum. This showed two low-energy peaks, presumably L x-rays and the 27-kev gamma ray (Fig. 23). Next, the gamma rays in coincidence with α_{110} were recorded, again showing L x-rays and a 27-kev gamma ray. The L x-ray peak appeared more intense in this run than in the previous run (see Fig. 23).

Figure 24 shows the result of the next experiment. The magnetic field was set so that α_{329} was the gate alpha group. In coincidence appeared K x-rays, the 145-kev Compton peak, and the 300-kev peak. The 300-kev peak appeared complex, and could be resolved into the same three components as before. Upon gating with α_{387} (Fig. 25), one observed K x-rays and a high-energy peak. The high-energy peak could be resolved into ~260, 300-, and ~360-kev components. The amount of 300-kev radiation, compared with the number of gamma rays just higher and lower in energy, was much less than in the previous α_{329}-gamma-ray experiment. The significance of these experiments is discussed further in the next section.

3. Decay Scheme of Protactinium-231

The basic skeleton for a decay scheme for this isotope is the set of energy levels populated by the alpha groups seen in the alpha-particle studies. The energies of the levels in the decay scheme in Fig. 26 are those derived from the alpha-particle studies (Table XVIII). The transitions shown there are from the data of the conversion-electron study of Falk-Vairant[50] and the numerous gamma-ray studies.

Table XIX gives a summary of the conversion-line data by Falk-Vairant. There the gamma-ray energies, conversion lines observed for each gamma ray, and the assigned multipolarities are listed. The assignments of the multipolarities of the various gamma rays were based on relative L-subshell ratios, where available, and K/L conversion ratios. The M2 and M1 assignments for the 331- and 301-kev gamma rays were confirmed by the gamma-ray studies reported here.

The 27.5-kev gamma ray has been assigned an E1 designation on the basis of its conversion coefficient.[50] In the delayed-coincidence

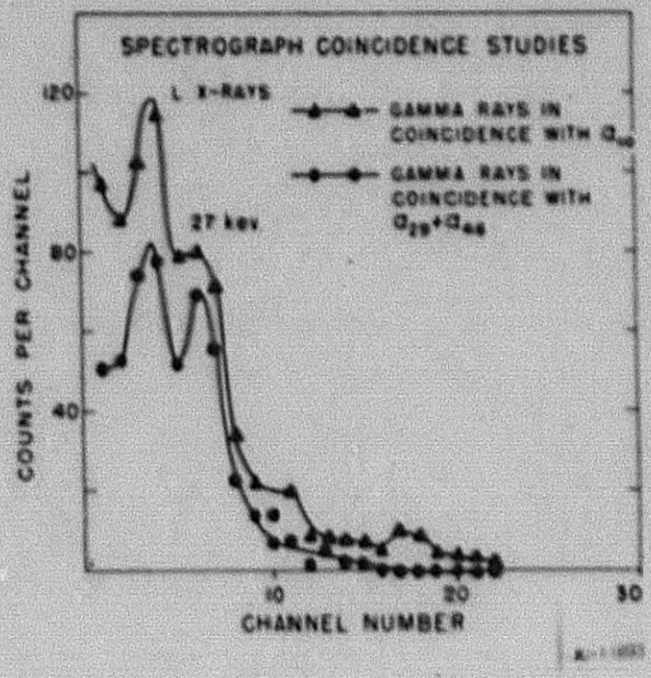

Fig. 23. Gamma-ray spectra in coincidence with α_{29} plus α_{46} and with α_{110} of Pa^{231}.

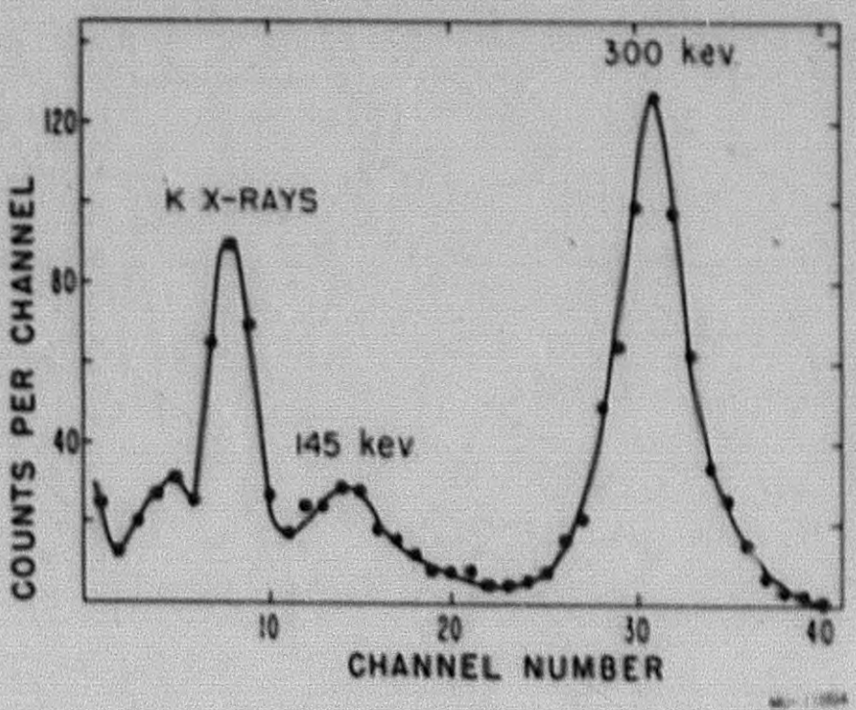

Fig. 24. Pa^{231} gamma-ray spectrum in coincidence with α_{329}.

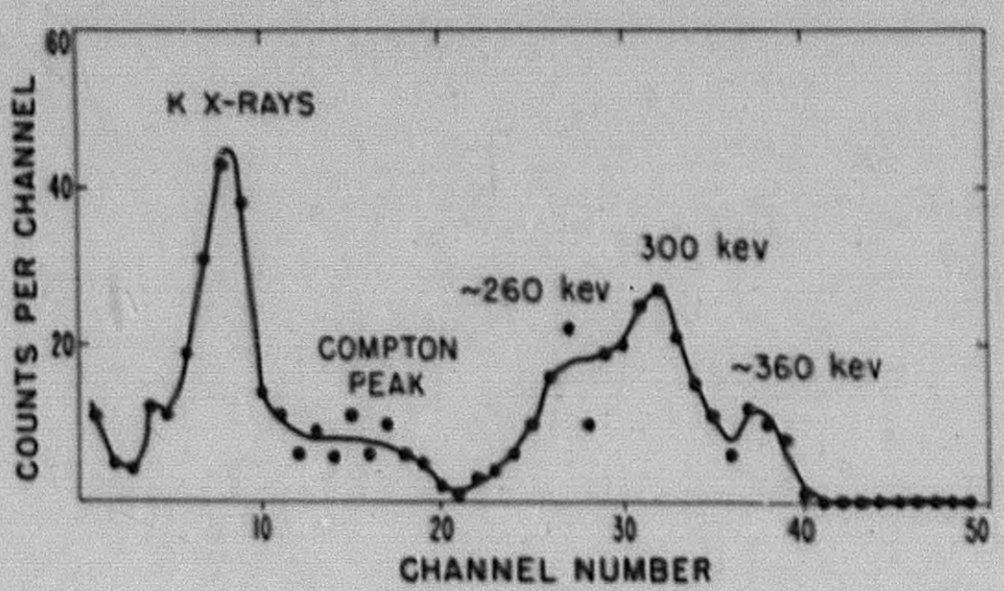

Fig. 25. Pa^{231} gamma-ray spectrum in coincidence with α_{387}.

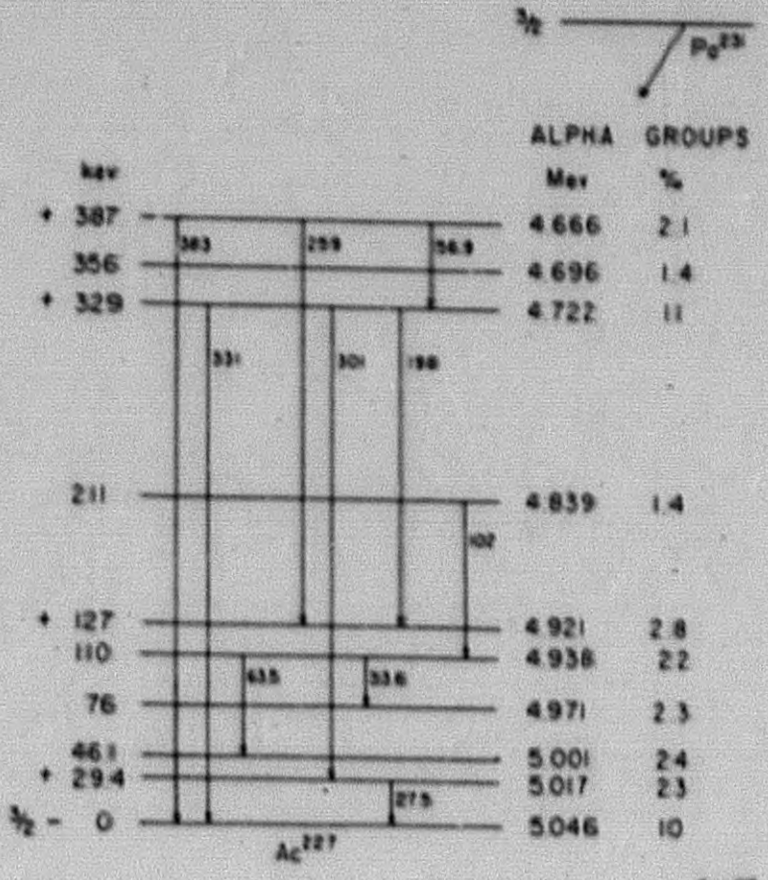

Fig. 26. Decay scheme of Pa^{231}

studies by Moore,[51] coincidences involving the 27-kev gamma ray always appeared delayed with but a few exceptions. However, the prompt coincidences involving the 27-kev gamma ray can be explained by assuming that the 27-kev radiation being detected is the iodine K x-ray associated with the escape-peak phenomenon. Thus, the 27-kev gamma ray is delayed, and Moore has measured its half life as 3.7×10^{-8} sec.

Table XIX

Results of conversion-electron study of Falk-Vairant on Pa^{231}

Gamma Ray (kev)	Observed lines	Multipolarity
383	K	E1 or M2
357	K, L	M1
331	K, L, M	M2
301	K, L, M	M1
259	K, L	M1
198	K, L	E2
102	L_{II}, L_{III}, M	E2
82.3	L_I, M, N	M1 or M2
63.5	L_{II}, L_{III}, M	E2
56.9	L_{II}, L_{III}, M	E2
38.0	L_I, L_{III}, M	E2
33.6	L_I, M	--

In the placement of these transitions into the level scheme described by the alpha-particle studies, a few trouble spots are found. First, from the specific alpha particle-gamma ray experiments, the 27-kev gamma ray must follow alpha decay to the 29.4- or 46.1-kev state. The fact that the 300-kev gamma ray is in coincidence with the 27-kev gamma ray places the 27-kev gamma ray as de-exciting the first excited state. However, the energy discrepancy between the 27.5-kev gamma ray and the 29.4-kev level separation from the alpha-particle studies is somewhat troublesome, since both these energies were obtained with very high precision. This difference might be considered as evidence for postulating a 2-kev transition, either preceding or following the 27.5-kev transition. There is no other evidence for such a transition, as the uncertainties in the electron energies are about 2%, and nothing significant can be concluded from them. Until more precise electron spectra are determined, it will be assumed that the 27-kev gamma ray does represent the transition from the first excited state to ground. All the transitions placed in the level scheme agree in energy (within the experimental uncertainties) with the separations obtained from the alpha-particle spectrograph studies. All the gamma rays reported by the electron study of Falk-Vairant (Table XIX) have been assigned except the 357, 82.3-, and 38.0-kev transitions. The 357-kev transition could be placed as either depopulating the 356-kev level to the ground state or depopulating the 387-kev level to the first excited state. The coincidence measurement of the gamma rays in coincidence with the 27-kev gamma ray does not rule out the possibility that a small amount of radiation above 300 kev is in coincidence. Thus, it is rather difficult to place the 357-kev transition in the decay scheme. The 82.3-kev transition could also be placed in two different positions, one from the 127-kev state to the 46-kev state, and one from the 110- to the 29-kev level. In either case, the agreement energywise between the gamma-ray and the alpha-particle separations is not exact. Therefore, the 82.3-kev transition has not been assigned. The 38.0-kev transition does not fit anywhere in the level scheme. Attempts to reassign its electron lines to other transitions have not met with success, either. It should

be pointed out that the assignments of the various transitions in the level scheme are consistent with the coincidence data discussed previously.

The partial decay scheme leaves a lot to be desired in many respects. Several levels (namely the 356-, 127-, 76- and 46.1-kev states) have no modes of de-excitation shown. The 356- and 127-kev levels may possibly involve two of the unassigned gamma rays just discussed. Also, it is not possible to assign spins to the many levels with any high degree of certainty. One can assign some relative parities, and this has been done in Fig. 26 for those states in which it is possible. The predominance of one type of parity is pretty evident.

One feature of the decay scheme bears mentioning, and that is the absence of any clearly defined rotational structure. Although several groups of levels appear with spacings similar to those seen in rotational patterns, the gamma-transition patterns do not confirm that they are indeed rotational bands. Also, the regular decrease in alpha population as one goes to higher members of a rotational band, as seen in the heavier region,[6] is not evident for any possible rotational bands here. The large degree of complexity of this decay scheme makes it look similar to that of Th^{227} (discussed in a later section), which also fails to show prominent rotational features.

The course of future work on this isotope should probably include an extension of the existing electron data. Certainly more precise measurements along this line would aid immeasurably in placing transitions into the decay scheme. Also, intensity data for the low-energy electrons (below 100 kev) are nonexistent at present, and work along this line would be useful. Probably the next most fruitful approach will be through extremely extensive specific alpha particle-gamma ray coincidence experiments utilizing a high-geometry alpha-particle spectrograph. The work of this type discussed in this presentation was meant to be preliminary in nature, and by no means could be called an extensive study.

E. Alpha Decay of Californium-246

Cf^{246} is an even-even alpha emitter. The interest in the alpha decay of this nuclide is to extend the developing systematics of the heavy even-even alpha emitters. In particular, the energy of the first excited state and the alpha-particle abundances to this state are of interest. Also, information concerning the decay to higher levels can be of value. A study of this nuclide in the alpha-particle spectrograph should easily give information on the decay to the first excited level, but it is not expected that decay to higher levels can be observed in the spectrograph with the amounts of activity presently available.

The preparation of Cf^{246} in a state of high radioactive purity can be effected by irradiating a mixture of curium isotopes ($Cm^{242-246}$) with alpha particles of about 40 Mev energy.[53] (The mixture of curium isotopes is obtained by prolonged neutron irradiation of Pu^{239}). Of the californium isotopes produced, Cf^{246} has a half life of 35.7 hr,[53] while Cf^{247}, Cf^{244}, and presumably Cf^{245} have half lives sufficiently short[53,54] to allow their complete decay during a period in which the Cf^{246} is not greatly reduced. No appreciable contribution to the radioactivity comes from Cf^{248} because of its relatively long half life (250 d)[55] and low yield, since there were only small amounts of Cm^{246} and Cm^{245} in the bombarded curium. Cm^{242} would be present as the daughter product of Cf^{246} decay.

The sample of Cf^{246} made available for this study contained initially 2.9×10^5 alpha disintegrations per minute. Since the transmission of the alpha-particle spectrograph is about 4×10^{-5}, it was necessary to make as long an exposure as possible in order to obtain a reasonable number of alpha tracks. The ratio of peak height to background was about 50 for the main group, which makes it impossible to measure alpha groups of abundance lower than several percent. From other spectra of even-even alpha emitters in the region, it was obvious that the transition to the ground state (0+) and first excited state (2+) should be discernible, but no others. The sensitivity of gamma-ray detection, however, is such that some information could reasonably be expected from this source on the expected transition to the second

excited state (44). At the time of the first measurements the Cm^{242} activity was small relative to the Cf^{246}. There was some Cf^{250} and Cf^{252} in the sample owing to their incomplete removal from the target curium before bombardment. The Cf^{250} and Cf^{252} contribute spontaneous fission-gamma ray coincidences. With the detection system used, the intensity of these coincidences is nearly constant over the energy range under consideration.

1. Alpha Spectrum of Californium-246

The Cf^{246} source was prepared by vacuum sublimation and exposed in the alpha-particle spectrograph. Two exposures (22.6 hr and 33 hr, respectively) were made before the sample became too weak. The spectrum (Fig. 27) so obtained showed, as expected, only two alpha groups characteristic of transitions to the ground state and first excited state of an even-even alpha emitter. One notes rather poor resolution on this run, with peak half widths on the order of 20 kev. Because of the rather small amount of activity available (in terms of the spectrograph geometry) and the 35.7-hr half life the exposure had to be made with the highest spectrograph geometry (1/8-inch slit system), which gives a corresponding poor resolution. A further increase in peak half width was also due to sample thickness. The primary concern in the vaporization process was a good yield (because of the small amount of activity available) rather than a uniformly thin sample.

In the first exposure, a total of 5,137 alpha tracks was registered, 78% in the main group and 22% in the group leading to the first excited state.

The energy difference between the two groups was found to be 42.0 kev. This energy difference is probably good to about 0.5 kev. When the correction is made for the recoil energy, the decay-energy separation (gamma-ray energy) is 42.7 kev. The appearance of the first excited state of Cm^{242} is also prominent in the β decay of Am^{242m}. This transition energy has been measured as 42.3 ± .2 kev with a bent-crystal photon spectrometer (Jaffe),[56] and from the

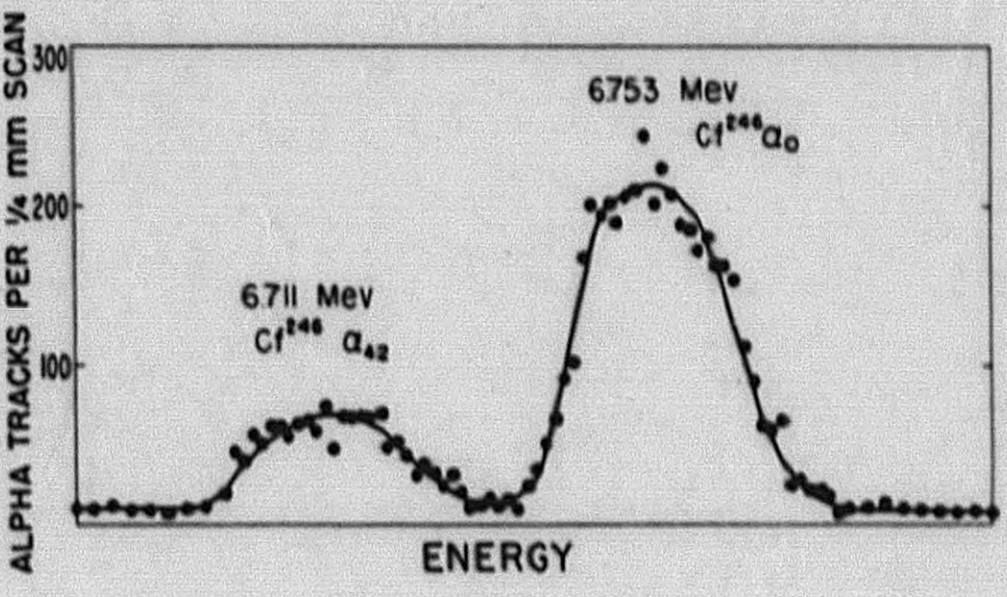

Fig. 27. Alpha-particle spectrum of Cf^{246}.

conversion-electron spectrum the values 42.2 ± .3 kev (Church)[57] and 42.12 ± .06 kev (Hollander et al.)[58] were obtained. The value obtained from the alpha-particle spectrum is in good agreement with these energies. This photon is also seen in the alpha-gamma coincidence spectrum of Cf^{246}, but the energy determination is not as accurate as the three values cited here.

In the second alpha spectrum taken, the photographic plate was also exposed to a source containing the U^{230} series. The Em^{218} line at 7.127 Mev[59] was taken as the energy standard, and from this the main group of Cf^{246} was found to be 6.753 Mev. When the recoil-energy correction is made, the decay energy for Cf^{246} becomes 6.865 Mev. The energy uncertainty here is rather large (± 10 kev) considering the uncertainty in the energy of the Em^{218} standard.

2. Alpha Particle-Gamma Ray Coincidence Spectrum

The gamma-ray spectrum in coincidence with the Cf^{246} alpha particles was measured with a scintillation spectrometer. Besides L x-rays, peaks at ~100 kev, ~60 kev, and ~44 kev showed up. The 100-kev photon is interpreted as the gamma ray leading from the sparsely populated second excited state to the first excited state. The 44-kev photon is the gamma ray from the first excited state to ground. The 60-kev radiation is probably due to Am^{241} impurity. Because of the high intensity of the Am^{241} 60-kev gamma ray and the low intensities of the Cf^{246} photons, an extremely small amount of Am^{241} (80 disintegrations per minute) would give the observed intensity. The L x-rays observed originate mostly from the L-shell conversion of the 44-kev transition.

The L x-ray abundance noted in the coincidence studies can be used as an indirect determination of the alpha abundance to the first excited state. This comes about from the fact that the first excited state drops to the ground state by an E2 transition, which is highly converted in the L shell. The method of making the coincidence measurement and corrections necessary to transfer these data into the degree of population to the first excited state are discussed elsewhere.[60] The alpha

population calculated in this manner was found to be 22%, in good agreement with the result obtained directly with the alpha-particle spectrograph (22%).

A few words are in order concerning the 44- and 100-kev gamma transitions. The conversion coefficient for the 44-kev gamma ray was determined from the ratio of the gamma-ray abundance and the previously discussed population to the 42-kev state (22%). Two determinations gave the L-shell conversion coefficient as 800 and 1200. These values are in the expected range for an E2 transition on the basis of extrapolations from the data of Gellman, Griffith, and Stanley.[40]

Four determinations gave an average intensity of 1.4×10^{-4} photon per alpha disintegration for the 100-kev transition. Since the energy of this photon corresponds well with the expected transition from the 4+ state to the 2+ state, we can assume that it is an E2 transition. Then if one estimates the conversion coefficient from others in this region, indicating a total conversion coefficient of about 18 for this energy, the intensity of alpha population to the 4+ state becomes 0.16%, as indicated in Fig. 28. Because of the approximations used in making this estimation, this probably represents a minimum alpha population to this level.

Asaro and Stephens[61] have re-examined the gamma-ray spectrum of Cf^{246} since the above work was published. In addition to 44- and 98-kev photons, a new gamma ray of 151 kev was observed. Since this energy is very close to that expected for the transition connecting the second and third excited states of an even-even nucleus in this region, this new gamma ray has been assigned that position in the decay scheme. From the abundance of the 151-kev gamma ray, the alpha population to the 6+ level is 1.5×10^{-2}%.

The alpha-particle and gamma-ray data just discussed are summarized in Fig. 28, which shows the decay scheme of Cf^{246}. The similarity to other even-even nuclei in this region is readily seen. A discussion of these results as they relate to the properties of the other even-even alpha emitters in this region is reserved for a later section.

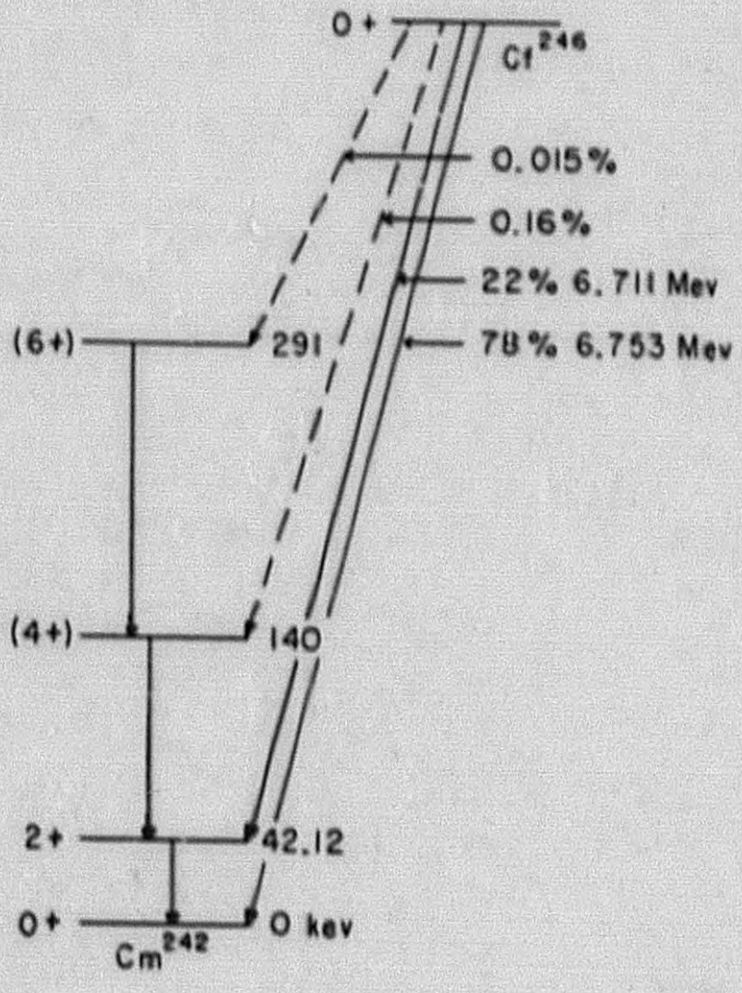

Fig. 28. Decay scheme of Cf^{246}.

F. Alpha Decay of Einsteinium-253

E^{253} is a beta-stable alpha emitter having a measured half life of 20 and 19.3 ± 0.3 days.[62,63] Various ion-chamber measurements of the alpha-particle energy of the main group have resulted in energies of 6.63 ± 0.02 Mev[62] and 6.61 ± 0.01 Mev.[63] In addition to the main group, Ghiorso et al.[64] have also found an alpha group at 6.25 Mev having an abundance of about 0.1%. These ion-chamber measurements represent the extent of the decay-scheme studies on this isotope before this work was begun. Stephens[10] has reported some preliminary experiments from the current work giving results from gamma-ray and alpha-particle studies and suggesting a decay scheme. Since then, the measurements on the alpha-particle spectrum have been continued, and indicate that a somewhat revised decay scheme is necessary.

This isotope is best prepared by pile irradiation of lower elements, ultimately uranium. All the samples used in the course of this work were prepared in this manner. The californium isotopes resulting from beta decay of the lower elements capture neutrons successively until the 20-day β^- emitter, Cf^{253}, is reached. This isotope beta-decays to E^{253} (20-day alpha emitter of interest here). The E^{253} undergoes a neutron capture reaction to give E^{254}, a 36-hour negatron emitter, which decays to Fm^{254}. Thus, upon purification, the einsteinium fraction contains predominantly two isotopes, E^{253} and E^{254}. E^{254} has two isomers, the 36-hour beta emitter and a one-year alpha emitter. In an irradiation of the sort just described, the E^{254} alpha activity is generally less by about a factor of 10^4 than E^{253} alpha activity. This ratio, of course, decreases as the E^{253} decays. Thus, one can essentially observe the E^{253} alpha spectrum in the total einsteinium fraction with very little interference from other isotopes. If a pure sample of E^{253} is desired (for gamma analysis, for instance), it can be obtained by milking it from the purified californium fraction.

This series of experiments was performed with three different E^{253} samples. They differed in the amount of activity owing to three factors, (a) the amount of starting material used in the reactor build-up, (b) the duration of neutron irradiation, and (c) the method of

sample preparation for the spectrograph. The first sample that was used was a vaporized sample (on platinum) containing 50,000 alpha disintegrations per minute. A second vaporized sample on platinum, of 99,000 alpha disintegrations per minute, was also used. The third sample contained 10^6 alpha disintegrations per minute and was prepared by electrolysis onto a gold foil. Owing to the small amounts of E^{253} activity available for the preparation of the vaporized samples, attempts were made to obtain the highest possible yield in the vaporization process. This resulted in the samples' being somewhat unevenly distributed over the platinum plates, which gave regions of fairly large thickness. The electroplated sample seemed to be of a much more uniform nature, and was exceptionally thin. These samples were all exposed in the alpha-particle spectrograph; the particulars of the exposures were as follows.

1. Alpha Spectrum of Einsteinium-253

Preliminary measurements to observe fine structure in the alpha decay of E^{253}, using the 50,000-disintegrations-per-minute sample, were unsatisfactory. The main source of difficulty was sample thickness, which caused a large amount of low-energy tailing on the peaks. The tailing made the observation of groups lower in energy and intensity than the main group rather difficult. However, tentative assignments of groups populating levels 43 kev and 90 kev above the level populated by the main group were made. The two low-intensity groups accounted for about 9% and 5% respectively of the decay. Also, the energy of the main transition was determined to be 6.642 Mev using the Em^{220} and Po^{216} alpha groups at 6.282 and 6.774 Mev[6] as standards. This energy is somewhat higher than the ion-chamber measurement quoted previously.

The data from these measurements were summarized by Stephens[10] and used along with his gamma-ray data to suggest a decay scheme for E^{253}. The main alpha group was considered to be the ground-state transition (a_0), and the two other groups were populating levels at 43 and 98 kev above the ground state.

The exposures made with the 90,000-disintegrations-per-minute sample also suffered some from the effects of sample thickness. The existence of two groups was clearly indicated, but the third group did not appear. A limit of 2% was set on its abundance, much less than the first experiments indicated.

The third sample was used for a series of experiments originally designed to check the existence of a third alpha group of E^{253}. Owing to two factors, this sample has given the best data to date on the E^{253} alpha fine structure. The fact that the counting rate was a factor of 10 higher than any of the previous samples coupled with the fact that an extremely good electrodeposition job yielded a uniformly thin deposit covering a small area (about 1/2 x 1/16 inches) made this sample ideal for considerable study. Eleven exposures were made with this sample. Some were designed to carefully study the most intense groups while others were designed to look for weak components of the spectrum. These two groups of measurements are discussed separately.

First of all a redetermination of the absolute energy of the main alpha group was made. Again, the E^{253} sample was exposed with a Th^{228} sample of comparable activity for 1 day. The Em^{220} and Po^{216} groups were used as standards, yielding 6.633 Mev as the energy of the E^{253} a_0. This measurement is much better than the previous result of 6.642 Mev because of much smaller half widths and a corresponding smaller uncertainty in the peak positions. The uncertainty in this energy is about 5 kev, from considerations of the uncertainties in the peak locations and energies of the standard groups.

The two main alpha groups, previously denoted as a_0 and a_{43}, were studied in a series of runs summarized in Tables XX and XXI. The energy separations are given in Table XX, where the exposure number, particle energy differences, and decay-energy separations are listed. The exposures listed there were made under widely varying conditions. The slit systems used with each exposure are also listed in Table XX. It would be expected that the exposure made with the 18-mil defining slit would be the most reliable energywise, and this has been considered in arriving at the best values in Table XX.

Table XX

Energy separation of E^{253} main alpha groups

Exposure No.	Slit width	Particle energy separation (kev)	Decay-energy separation (kev)
470	1/8 inch	41.3	42.0
472	1/16 inch	41.4	42.1
473	18 mil	40.9	41.6
476	1/8 inch	41.3	42.0
478	1/8 inch	41.3	42.0
480	1/16 inch	41.3	42.0
481	1/16 inch	41.4	42.1
	Best values	41.2 ± 0.6 kev	41.9 ± 0.6 kev

The peak half width on Exposure 473 was 7 kev. The energy uncertainty listed in Table XX considers both the uncertainty in locating the peaks on the photographic plate and a 1% calibration uncertainty. As the best value for the decay-energy separation between these two groups is 41.9 kev, the second alpha group is called α_{42}. Figure 29 shows the spectrum obtained in one of these runs (Exposure 473).

The abundance data from these runs are summarized in Table XXI. In arriving at the best value of 0.085, the runs on which the largest number of tracks were registered were weighted most heavily. The uncertainty listed in Table XXI is just the average deviation of the values in Table XXI. If no other alpha groups existed in the E^{253} spectrum, the abundances of α_0 and α_{42} would be 92.2% and 7.8% respectively. The next few paragraphs show that other alpha groups do exist in this spectrum, however.

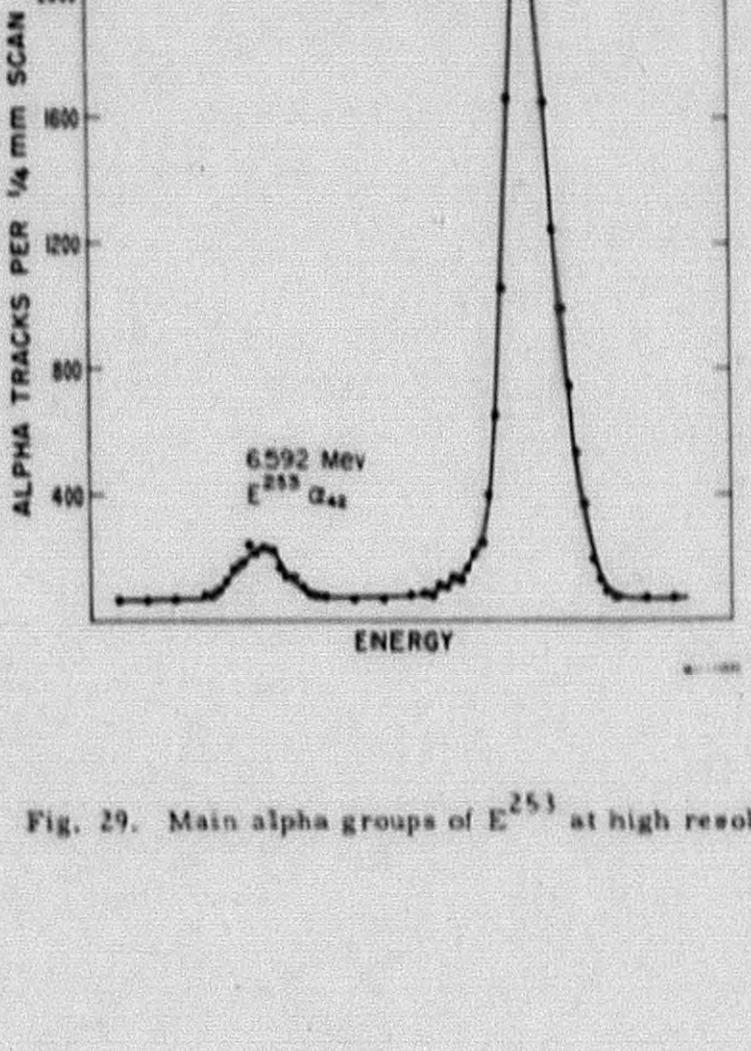

Fig. 29. Main alpha groups of E^{253} at high resolution.

Table XXI

Abundance ratio of the main E^{253} alpha groups

Exposure No.	α_{42}/α_0 abundance ratio
470	0.086
472	0.073
473	0.069
476	0.094
478	0.091
480	0.082
481	0.087
Best value	0.085 ± .007

Several other exposures were made with this sample, to study any low-intensity components of the E^{253} spectrum. The first of these is shown in Fig. 30. There two other groups of low intensity are seen. The data collected from these runs regarding the two low-abundance peaks are given in Tables XXII and XXIII.

Table XXII gives the data on the energies of the two groups. All the exposures listed there were run under high-geometry conditions. Since these groups are in low intensity, it would be expected that precise measurements of their energies would be extremely difficult. A look at Table XXII shows that there is a rather wide range in energies for each alpha group, which is borne out by the somewhat large uncertainties attached to these measurements. Because Exposures 474 and 479 were shorter than the others, the fourth alpha group was just barely detectable in those two runs. In arriving at the best value for the energy separation of the alpha group closest to α_{42}, the various runs were weighted according to the peak height of the new group. After the recoil correction is made, the corresponding decay-energy separations are 48.1 kev

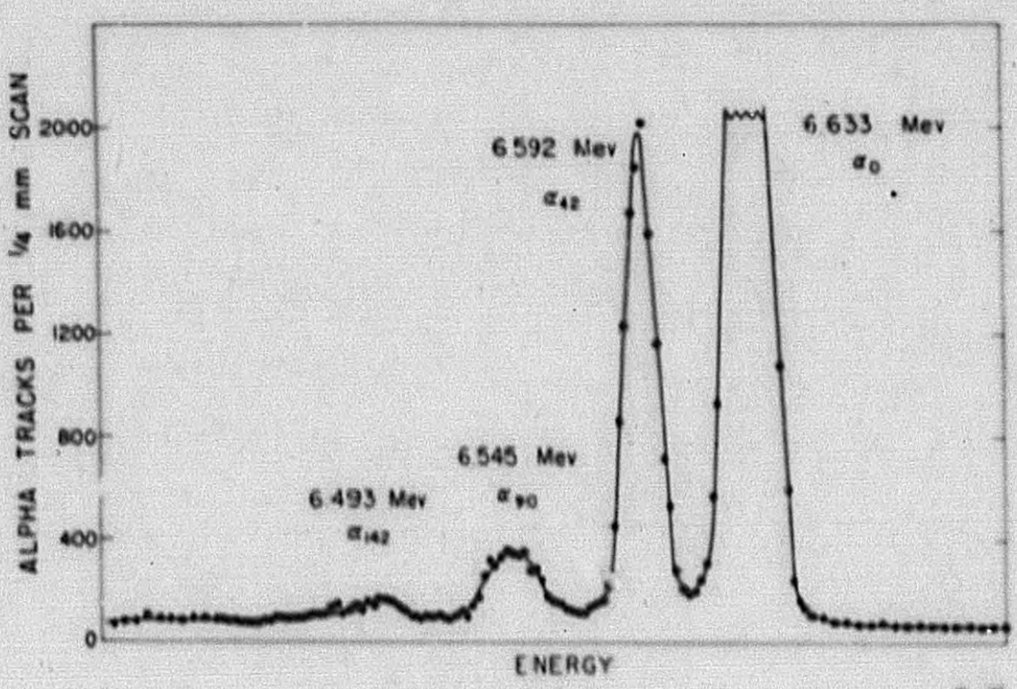

Fig. 30. E^{253} alpha-particle spectrum showing the low-intensity groups.

Table XXII

Low-intensity alpha groups of E^{253} energy data

Exposure No.	α_{42}-α_{90} separation (kev)	α_{42}-α_{142} separation (kev)
471	47.3	99.7
474	45.5	---
477	48.6	97.5
479	46.4	---
Best values	47.3 ± 1.5 kev	98.6 ± 2.0 kev

and 100.2 kev. As these are the decay-energy separations from α_{42}, the new alpha groups populate levels at 90.0 and 142.1 kev and are called α_{90} and α_{142}.

The abundance data from the same exposures are summarized in Table XXIII. Again, for arriving at the best values, the runs were weighted according to the number of tracks recorded.

The results from Tables XX to XXIII are summarized in Table XXIV. There, the absolute energy, decay energy to α_0, percentage abundance, and partial alpha half life (based on 19.3 days for the E^{253} half life) are given for each alpha group. Also included in this table is the low-intensity group reported by Ghiorso. Its decay-energy separation from the ground state has been fixed by Stephens' gamma-ray work as 393 kev. The limit of detection in the work reported herein was such that a group of as low an intensity as α_{393} (0.05% to 0.1%) would not have been detected.

2. Decay Scheme of Einsteinium-253

Before the decay scheme is discussed, the results of some gamma-ray studies of E^{253} are given. Stephens[10] found L x-rays and gamma rays of 43, 56, 112, and 393 kev. The 112-kev radiation is probably berkelium K x-rays. The intensities of the observed radiations were 5.3% (L K-rays),

Table XXIII

Low-intensity alpha groups of E^{253} abundance data

Exposure	α_{90}/α_{42} Abundance ratio	α_{142}/α_{42} Abundance ratio
471	0.231	0.047
474	0.216	----
477	0.255	0.039
479	0.222	----
Best values	0.235 ± .014	0.043

Table XXIV

E^{253} alpha fine structure

Energy (Mev)	Decay energy to α_0 (kev)	Abundance (%)	Partial alpha half life (days)
6.633	0	90.2	21.4
6.592	41.9	7.7	250
6.545	90.0	1.8	1100
6.493	142.1	0.3	6000
6.246	393	0.05 - 0.1	$\sim 3 \times 10^4$

0.09% (43-kev gamma ray), 0.02% (56-kev gamma ray), 0.03% (K x-rays), and 0.04% (393-kev gamma ray).

Asaro[65] has studied the low-energy gamma radiations of E^{253} with a proportional counter by observing the escape peaks of the gamma rays. He reports a gamma ray of 41.7 ± 0.3 kev.

A decay scheme for E^{253} is given in Fig. 31. It is seen that the gamma ray reported by Asaro falls readily into place. The 43-kev gamma ray reported by Stephens is probably the transition depopulating the first excited state, i.e., the same gamma as the 41.7-kev gamma ray reported by Asaro. The 56-kev gamma ray reported by Stephens may be the transition between the third and second excited states.

Using the abundance of the 43-kev gamma ray (0.09%) and considering that all of the alpha population to the 90- and 142-kev levels is de-excited through the 42-kev level, one calculates a conversion coefficient of 110 for the 43-kev gamma ray. This seems to indicate predominantly M1 or E2 character for this transition. Stephens has concluded that the 393-kev transition is M1 or M2.

Since the analysis of the gamma-ray data has given little satisfaction as to conclusions on the multipolarity of the transitions, one is somewhat free to speculate about the nature of the levels seen in this decay. The grouping of close-lying levels having regular spacings between them and regular alpha-particle intensity patterns has been considered to be characteristic of rotational bands in this region.[6] If the lowest levels in Bk^{249} do belong to a rotational band, they may be expected to follow the I (I+1) energy rule, where I is I_0 (spin of the base level of the band), $I_0 + 1$, $I_0 + 2$, etc. Taking the first two energy spacings, one can calculate I_0. In this case, the equations to consider are

$$C\left[(I_0 + 1)(I_0 + 2) - (I_0)(I_0 + 1)\right] = E_{1-0},$$

$$C\left[(I_0 + 2)(I_0 + 3) - (I_0 + 1)(I_0 + 2)\right] = E_{2-1},$$

which reduce to

$$\frac{E_{2-1}}{E_{1-0}} = \frac{(I_0 + 2)}{(I_0 + 1)}.$$

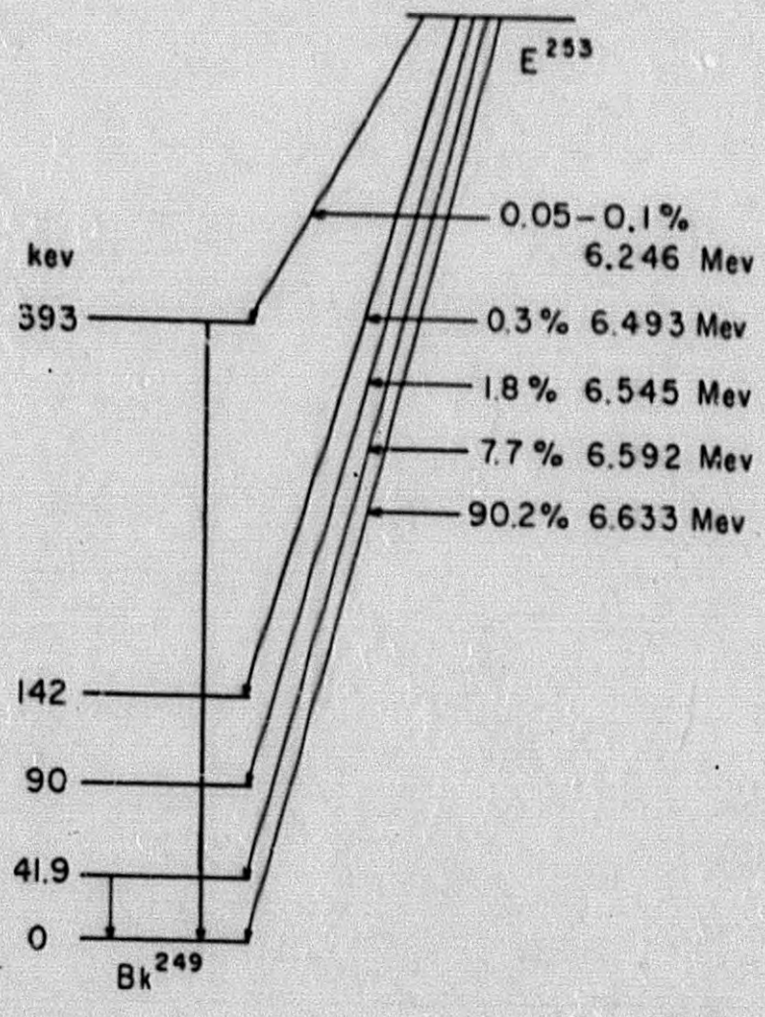

Fig. 31. Decay scheme of E^{253}.

If we take the energy separations of 41.9 and 48.1 kev from the alpha-particle data, I_0 is 5.8, indicating an 11/2 spin for the ground state of Bk^{249}. Using this as a basis, one expects the spacing between the second and third excited states to be 54.7 kev, which is in fair agreement with the separation observed in the alpha-particle study. If one considers the energy uncertainties on these separations, the calculation above yields an I_0 varying from 9/2 to 15/2. This, at any rate, indicates a rather high spin for the ground state of Bk^{249}. Some of the aspects of this proposed rotational band are discussed in a later section.

G. Alpha Decay of Plutonium-236

The isotope Pu^{236} is a 2.7-year[66] alpha emitter whose alpha-particle energy has been measured as 5.75 Mev by an ion-chamber technique. This energy is the energy of the main alpha group in this decay, distorted to some extent because of unresolved fine structure. The only study of the decay scheme of this isotope was that by Dunlavey and Seaborg,[67] who observed alpha-particle and conversion-electron tracks in emulsions saturated with Pu^{236}. They reported that 20% of the alpha decays populated a level about 45 kev above the ground state of U^{232}.

A sample of Pu^{236} for this decay-scheme study was made by milking it from Np^{236}. The Np^{236} was made by bombarding U^{235} with deuterons in the Crocker Laboratory 60-inch cyclotron. The Np^{236} decays with a 22-hour half life (33% negatron branching) to give Pu^{236} and U^{236}. The plutonium made in this way is composed almost exclusively of Pu^{236}, since Np^{236} is the only negatron emitter that is produced in the bombardment. The plutonium was purified, and an electroplated source prepared which was suitable for exposure in the alpha-particle spectrograph. The sample was somewhat thick, causing a certain amount of low-energy tailing. There were about 2×10^6 alpha disintegrations per minute in the sample, so that high-resolution studies on the main fine-structure components were possible. Any low-intensity components would be discernible only with low resolution-high geometry experiments.

1. Main Alpha Groups of Plutonium-236

From the work by Dunlavey and Seaborg, one would expect to observe two intense groups in the Pu^{236} alpha spectrum. A series of exposures was made to examine these two groups. The results from this series are given in Tables XXV and XXVI. All the exposures listed there were made with the 18-mil defining slit arrangement (high-resolution condition), and were run for about the same length of time (9 hours). A typical spectrum is shown in Fig. 32, in which two prominent groups are apparent. Table XXV lists the energy separations obtained with these exposures, and Table XXVI summarizes the abundance data. The energy uncertainty in Table XXV includes a 1% uncertainty in the spectrograph calibration.

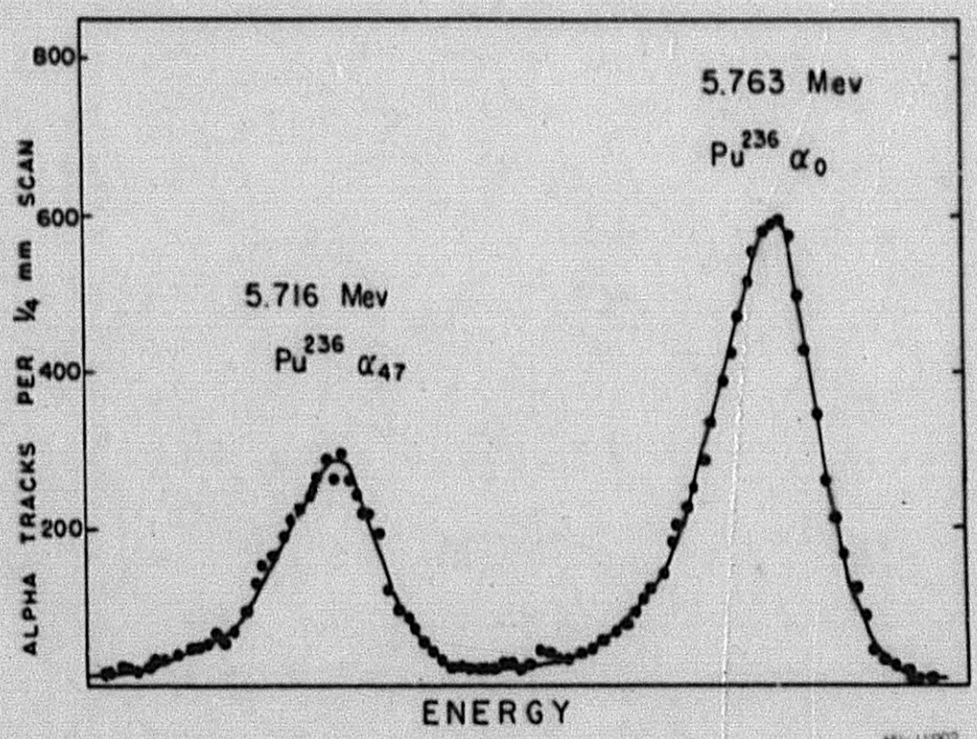

Fig. 32. Main components of Pu^{236} alpha-particle spectrum.

An exposure designed to determine the absolute energies of the Pu^{236} alpha groups was made using a Th^{228} sample along with the Pu^{236} sample. The Ra^{224} (5.681 Mev) and Em^{220} (6.282 Mev)[6] alpha groups were used as standards, giving 5.763 Mev as the energy of the main Pu^{236} group. This is slightly higher than the 5.75-Mev value obtained with an ion chamber. One would expect the ion-chamber value to be lower because it would be influenced by the unresolved fine structure.

From the systematics of even-even emitters,[6] one would expect that the highest-energy alpha transition populates the ground state of the U^{232} nucleus. Thus, the highest-energy group is called α_0. As the lower-energy alpha group populates a level 47.5 kev above the ground state, it is called α_{47}.

If no other alpha groups exist in this decay, the abundance ratio in Table XXVI shows intensities of 69.0 ± 0.5% and 31.0 ± 0.5% for the two alpha groups, α_0 being the most prominent. These abundance data show a larger population to the first excited state than was indicated by the earlier, less sensitive emulsion study. The abundances just mentioned are revised somewhat after the low-intensity alpha group is discussed in the next section.

2. Low-Intensity Alpha Group of Plutonium-236

On considering the complex alpha-particle spectrum of the other even-even plutonium isotopes, one sees that there is a good chance that Pu^{236} would have another alpha group in abundance of about 0.1%. Both Pu^{240} and Pu^{238} are known to have a third alpha group of this intensity,[6] and in both cases it populates a level at about 150 kev. If such a group exists in Pu^{236} decay, and if its intensity is ~0.1%, it should be discernible in high-geometry runs with the source used for the experiments just described. Therefore, two exposures were made in a search for a third component of this spectrum.

Table XXV

Main alpha groups of Pu^{236} - energy data

Exposure No.	Particle-energy separation (kev)	Decay-energy separation (kev)
447	46.4	47.2
451	46.8	47.6
452	46.9	47.7
Best values	46.7 ± 0.6 kev	47.5 kev

Table XXVI

Main alpha groups of Pu^{236} - abundance data

Exposure No.	a_{47}/a_0 abundance ratio
447	0.435
451	0.454
452	0.460
Best value	0.450 ± .010

Because of the size of the sample available and the expected low intensity of a third group, the exposures were 5 and 6 days in length, and a 1/16 - inch defining slit was used. As expected, a third group did appear at the expected energy. The data from the two exposures in this series are given in Table XXVII. Figure 33 is one of the spectra obtained in this search.

The two exposures listed in Table XXVII were run 75 days apart under identical conditions. Since there was a very large number of alpha tracks in α_0 and α_{47}, they were not scanned completely on the long runs. To determine the positions of these peaks on the photographic plate, one field of view was counted for every millimeter in the horizontal direction. To determine the abundance of the new group, the number of tracks in α_0 and α_{47} was determined by running a much shorter exposure under an identical source setup and then calculating the number of tracks on the long run from those recorded in the shorter one.

After correction for the recoil energy, the decay-energy separation between the new group and α_0 is 155.9 kev; thus the new low-intensity alpha group in Pu^{236} decay is denoted as α_{156}.

Table XXVIII summarizes the present data on the alpha-particle spectrum of Pu^{236}. There, the absolute energy, the decay energy to α_0, the abundance, and the partial half life are given for each group. The partial half lives are based on a 2.7-year half life for Pu^{236}. A limit of 0.07% has been set on the existence of other alpha groups down to 300 kev below the energy of α_0 (Exposure 450).

3. Gamma Rays of Plutonium-236

A sample of 10^6 alpha disintegrations per minute was chosen for a study of the gamma radiation in the decay of Pu^{236}. An alpha pulse-height analysis showed this sample to be 97.5% Pu^{236} with Pu^{238} accounting for the rest of the activity. The Pu^{238} presumably came from U^{238} present in the uranium that was originally bombarded. The daughter activities in this sample at the time of the experiment were present in about 0.1% abundance for each member of the chain.

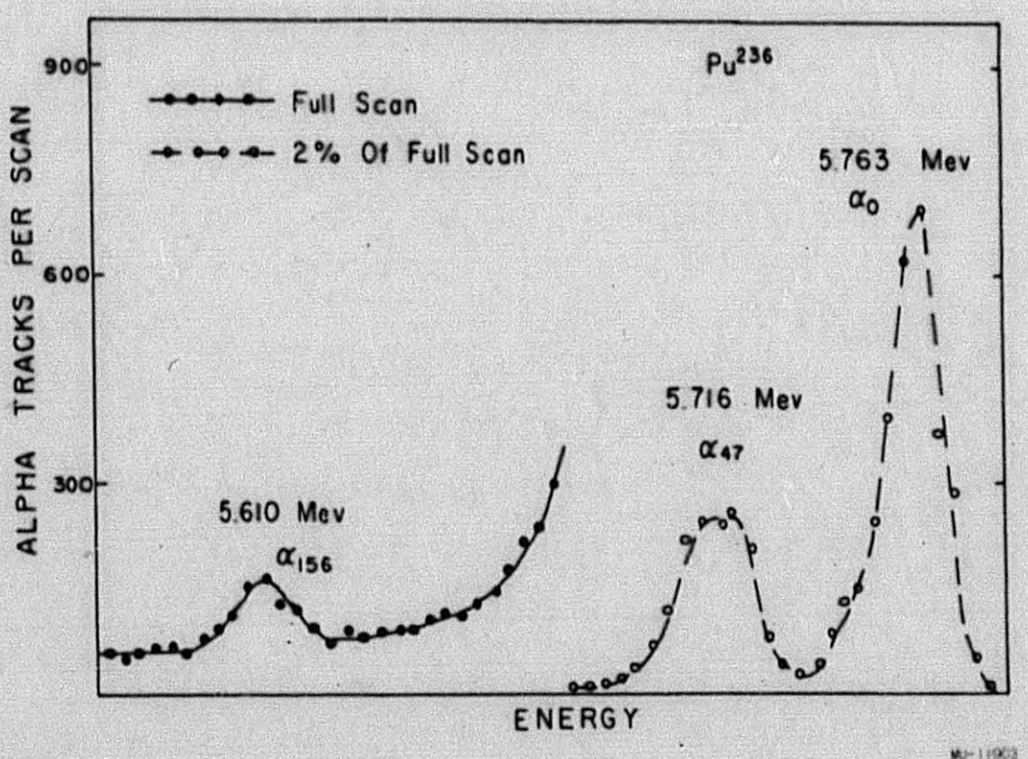

Fig. 33. Pu^{236} alpha spectrum showing the low-intensity group.

Table XXVII

Low-intensity group of Pu^{236}

Exposure No.	a_{156}-a_0 energy separation (kev)	a_{156} abundance (%)
450	153.5	0.20
484	153.0	0.16
Best values	153.3 kev	0.18%

Table XXVIII

Alpha-particle spectrum of Pu^{236}

Alpha group	Absolute energy (Mev)	Decay energy to a_0 (kev)	Abundance (%)	Partial alpha half life (years)
a_0	5.763	0	68.9	3.9
a_{47}	5.716	47.5	30.9	8.7
a_{156}	5.610	155.9	0.18	1500

The gamma spectrum obtained by running alpha particle–gamma ray coincidences with this sample is shown in Fig. 34. The low-energy peak is probably due to L x-rays, and the peak at ~65 kev is undoubtedly due to platinum K x-rays. This sample was mounted on 1-mil platinum, and the gamma rays were detected through the platinum. Thus, the L x-ray peak has been attenuated a great deal. The gamma rays of 47, 110, and 165 kev have been assigned to Pu^{236}. The intensities of these gamma rays are 3.1×10^{-4}, 1.2×10^{-4}, and 6.6×10^{-6} photons per alpha particle respectively.

A search was made for higher-energy gamma rays in this sample by running alpha particle–gamma ray coincidences. After subtraction out of the chance coincidence contribution, several very low-intensity peaks remained in the energy range of 200 to 900 kev. Because of the very poor statistics on this run, it is rather difficult to determine if any of these peaks are due to real gamma rays. At the intensities being dealt with here, gamma rays from the daughter activities in the sample might interfere to a large extent.

4. Decay Scheme of Plutonium-236

A decay scheme for Pu^{236} alpha decay is shown in Fig. 35. The 47-kev gamma ray undoubtedly depopulates the first excited state. The 110-kev gamma ray fits very nicely as the transition from the second excited state to the first. By analogy with other even-even nuclei in this region,[6] the 165-kev gamma ray has been placed as decaying from a level not seen in the alpha-particle work to the second excited state.

Using the alpha population to the first excited state (30.9%) and the intensity of the 47-kev gamma ray (3.1×10^{-4}), one calculates a conversion coefficient of 1000 for this gamma ray. This is about what one would expect for an E2 transition of this energy on the basis of extrapolations from the data of Gellman, Griffith, and Stanley.[40] As the ground state of U^{232} is presumed to be 0+, the first excited state is thus 2+.

From the alpha population to the second excited state (0.18%) and the intensity of the 110-kev gamma ray (1.2×10^{-4}), a conversion coefficient of 14 is calculated for the 110-kev gamma ray. This would be

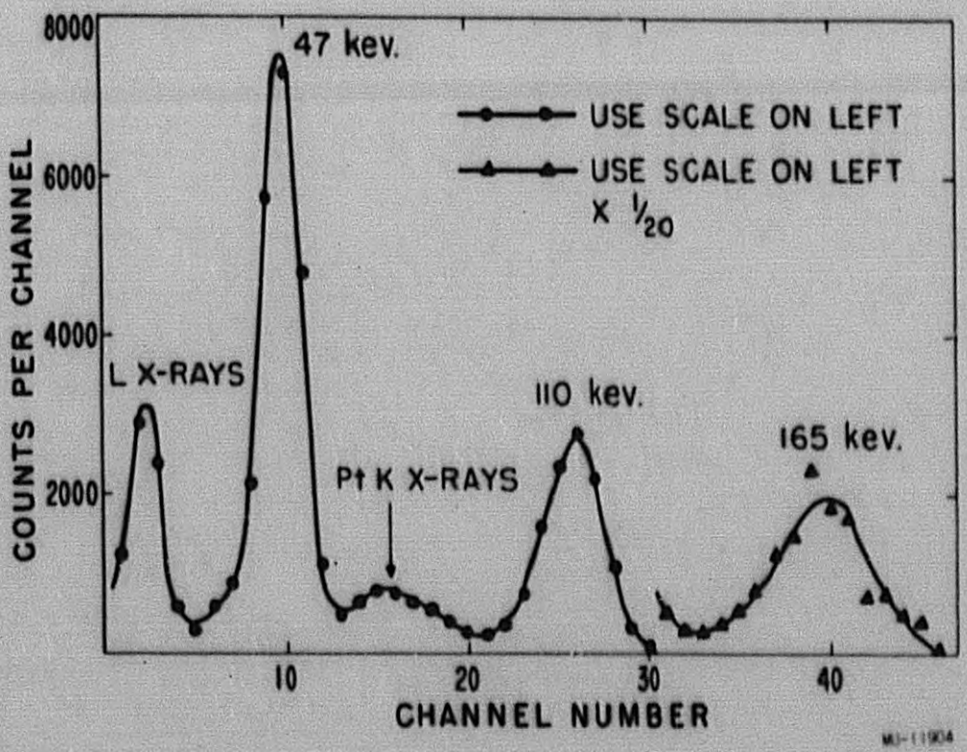

Fig. 34. Pu^{236} alpha particle–gamma ray coincidence spectrum.

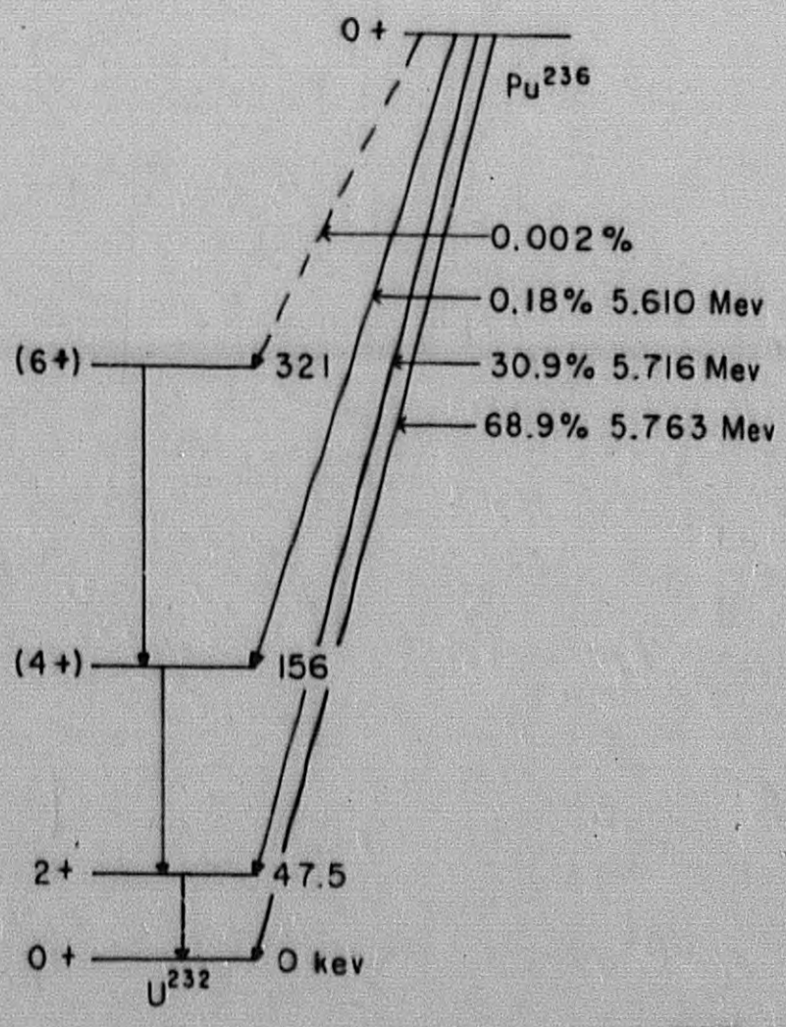

Fig. 35. Decay scheme of Pu^{236}.

consistent with an E2 designation. Thus, the first two excited states seem to conform to the pattern predicted by the collective model.[42] If we accept this premise and assume that the 165-kev gamma ray is the transition between the 6+ and 4+ members of a rotational band, this transition should be E2 in nature and have a conversion coefficient of about 2. This would require an alpha population to the 6+ level of 2×10^{-5}, which is well below the sensitivity of these measurements.

Ong Ping Hok[68] has studied the beta decay of Pa^{232} and finds that levels at 47.2 and 156.3 kev are populated in U^{232}. These energies are in very good agreement with those from the alpha-particle work for the energies of the first two excited states. On the basis of relative L-shell conversion coefficients, he has assigned E2 designations to the gamma rays de-exciting these levels. This is consistent with the conversion coefficients just discussed. He has also reported a 175.3-kev gamma ray, which is not in good agreement with the 165-kev energy measured in the gamma spectrum in Fig. 34. Pa^{232} appears to populate several other levels at higher energy. There is no evidence as yet for population of these levels in the alpha decay of Pu^{236}.

A well-developed rotational band pattern is quite evident in the decay of Pu^{236}. Some of the aspects of this pattern, especially the alpha populations to the various levels, are discussed later.

H. Alpha Decay of Actinium-225

Ac^{225} is a beta-stable alpha emitter with a half life of 10.0 days.[43] It is a member of the Th^{229} decay family, and this relationship was exploited as a source of the Ac^{225} activity. Previous studies on the decay properties of this isotope are limited to an ion-chamber measurement of its alpha-particle energy,[69] a study of the alpha-particle and gamma-ray spectra reported in 1955 by Stephens[10] of this laboratory, and a recent conversion-electron study by Stephens and Asaro.[70] Although Stephens observed four alpha groups in the fine structure of this decay, the resolution of these components was such as to merit a re-investigation of the alpha spectrum.

The source of the Ac^{225} used in the study described here was a sample of 10^7 alpha disintegrations per minute of Th^{229}. This Th^{229} was originally prepared by milking it from its U^{233} parent. The U^{233} was prepared by neutron irradiation of Th^{232} to give Th^{233}, which yields U^{233} after two successive beta decays. The Th^{229} milked from this U^{233} contained appreciable quantities of Th^{232} and Th^{228}, presumably from an incomplete initial separation of uranium from thorium. The Ac^{225} results from the beta decay of Ra^{225}, which is the alpha daughter of Th^{229}. The Ac^{225} was separated from the equilibrium mixture by an extraction precedure described by Stephens in the earlier work.[10] A small percentage of the radium carried over into the actinium fraction. This would present no problem at all for the alpha-particle spectrum study if the radium were all beta-emitting Ra^{225} formed from the Th^{229} decay. The Th^{228} present in the thorium sample has Ra^{224} in equilibrium with it. Ra^{224} is an alpha emitter having an alpha group near the Ac^{225} groups,[71] which would complicate the observed spectrum. However, by observing the rate of decay of any alpha groups found in the spectrum, one can easily make a definite assignment to Ac^{225} or Ra^{224}. This sample also contains the daughters of Ac^{225}, but these should not interfere in the alpha-particle measurements, since their alpha-particle energies are all well above that for Ac^{225}.[43]

A sample containing ~10^6 alpha disintegrations per minute of Ac^{225} was prepared for spectrograph use by vacuum sublimation onto a 2-mil platinum plate. This sample was then used for four exposures on the alpha-particle spectrograph, which is discussed next.

1. Alpha Spectrum of Actinium-225

The four alpha groups reported by Stephens[10] in the decay of Ac^{225} were at 5.815 Mev (54%), 5.779 Mev (29%), 5.717 Mev (13.5%), and 5.628 Mev (3.5%). The two most intense groups are separated by only 36 kev, and they were not well resolved.

The first of the four runs made in this study was made for 2 days with a 1/8-inch-wide slit system masking the sample (maximum-geometry condition). As expected, the spectrum shown in Fig. 36 is very similar to that obtained by Stephens, with two exceptions. The first is the improved resolution of the main groups, and the second is the presence of a fifth alpha group between the two lowest-energy groups previously reported for Ac^{225}. This is in the approximate position expected for a group of Ra^{224} known to be present in the sample (5.681 Mev), and a tentative assignment to Ra^{224} was made for the fifth alpha group. The data regarding the energy separations and abundances of the Ac^{225} alpha groups from this and succeeding runs are summarized in Tables XXIX and XXX.

In an attempt to obtain the best resolution possible with this sample, a 5-day exposure was made with an 18-mil defining slit. The spectrum observed in this run is shown in Fig. 37. One notes almost complete resolution of the two most intense groups. Also, the group tentatively assigned to Ra^{224} has decayed relative to the Ac^{225} groups. The amount of decay is consistent with the known half lives of the two isotopes (Ac^{225}, 10 days; Ra^{224}, 3.6 days).[43] Also, a peak of small abundance (indicated by the arrow in Fig. 37) has appeared. This group would not have been discernible on the previous low-resolution run. This new group is tentatively assigned to Ac^{225} decay.

A third exposure under the same conditions as the first (1/8-inch slit) was made for 4 days to confirm the assignment of the one group to Ra^{224}. This spectrum was essentially the same as Fig. 36 except that

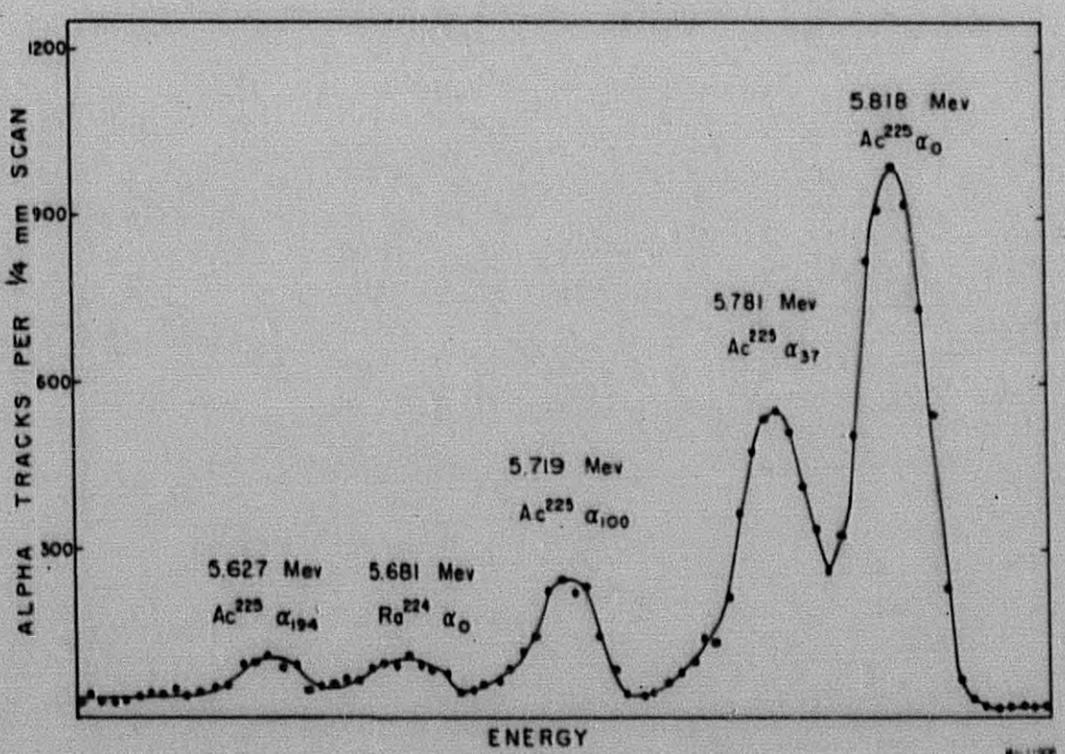

Fig. 36. Ac^{225} alpha-particle spectrum at low resolution.

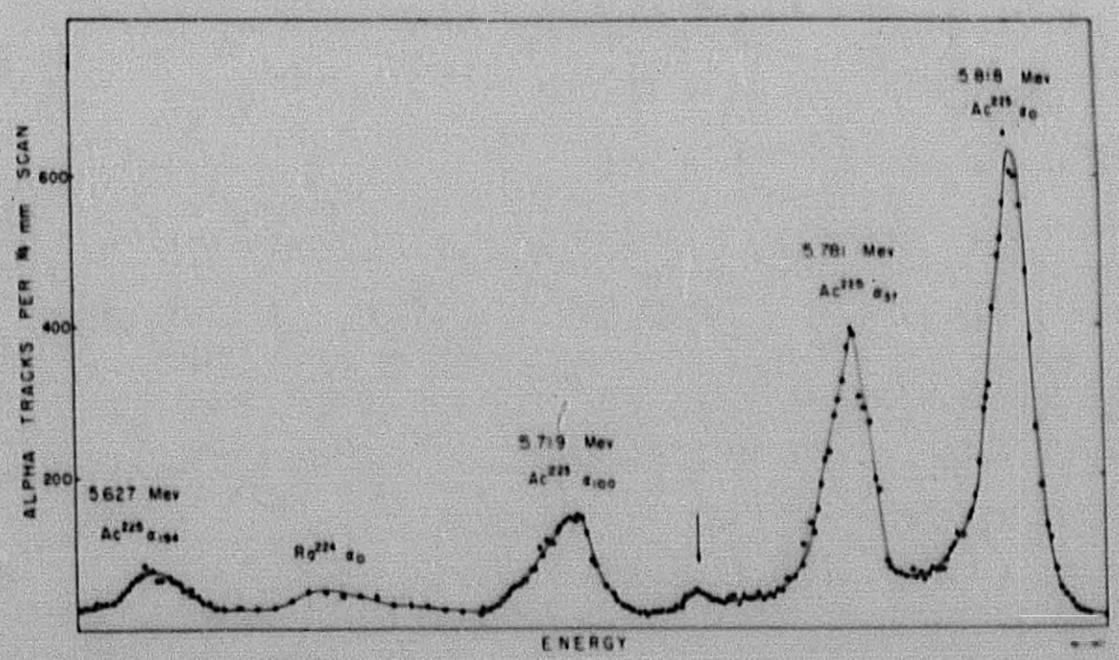

Fig. 37. High-resolution alpha-particle spectrum of Ac^{225}.

the group assigned to Ra^{224} has decayed relative to the Ac^{225} by an amount expected from the half lives involved.

The fourth exposure in this series was made with a 1/16-inch-wide slit. Besides the Ac^{225}, a sample containing the Ra^{226} family was also mounted in the source holder. The Ra^{226} daughter alpha groups at 5.486 Mev (Em^{222}) and 5.998 Mev (Po^{218})[6] were used as energy standards. The energy of the highest-energy (most abundant) Ac^{225} group was 5.818 Mev from this measurement. The previous measurements of 5.80 Mev[69] and 5.815 Mev[10] are in good agreement with this value. The uncertainty in both spectrograph measurements is fairly large (8 to 10 kev), considering the corrections that had to be made to account for the different half widths (due to different sample thicknesses) of the standard and Ac^{225} groups.

It should be pointed out that Stephens in his earlier work was not troubled by Ra^{224} in his Ac^{225} sample. In his studies, the radium fraction was separated and allowed to decay for a month before the measurements were made. This length of time would be sufficient for the Ra^{224} to decay almost completely.

The data from this series of exposures are summarized in Tables XXIX and XXX. The energy separations from the highest-energy group are given in Table XXIX. It would be expected that the high-resolution run (446) would yield the best separations, and this has been considered in determining the best values. As is evident from Stephens' gamma-ray data, the highest-energy alpha transition is considered the ground-state transition. After correction for the recoil energy, the other alpha groups are found to populate levels 37.2, 72, 101, and 194 kev above the ground state, and are called α_{37}, α_{72}, α_{101}, and α_{194}. The abundance data are summarized in Table XXX. Again the highest-resolution run is expected to give the best data, and this has been considered in arriving at the best values.

Table XXXI summarizes the complex alpha spectrum of Ac^{225}. The absolute energy, decay-energy separation from α_0, abundance, and partial alpha half life are given for each group. The partial half lives are based on the half life of 10.0 days[43] for Ac^{225}. It should be reemphasized that the assignment of the low-intensity group at 5.747 Mev to Ac^{225} is still tentative.

Table XXIX

Energy separations of Ac^{225} alpha groups

Exposure No.	Particle energy separation from α_0 (kev)			
	α_{37}	α_{72}	α_{101}	α_{194}
445	36.0	--	99	189
446	36.6	71	100	191
448	35.9	--	99	190
449	37.2	--	97	192
Best values (kev)	36.5	71	99	191
Decay-energy separation from α_0	37.2	72	101	194

Table XXX

Abundances of Ac^{225} alpha groups

Exposure No.	Abundance (%)				
	α_0	α_{37}	α_{72}	α_{101}	α_{194}
445	57.2	28.1	--	10.7	4.0
446	54.8	27.5	0.8	12.2	4.7
448	54.8	29.9	--	11.9	3.4
449	54.6	26.8	--	14.7	3.9
Best values	55.1	27.9	0.8	12.1	4.1

Table XXXI

Complex alpha spectrum of Ac^{225}

Energy (Mev)	Decay energy to α_0 (kev)	Abundance (%)	Partial alpha half life (days)
5.818	0	55.1	18.1
5.781	37.2	27.9	35.8
5.747	72	0.8	1300
5.719	101	12.1	82.6
5.627	194	4.1	250

2. Decay Scheme of Actinium-225

A suggested decay scheme for Ac^{225} is given in Fig. 38. It is essentially the same as the one proposed by Stephens except for some minor modifications coming from the improved alpha-particle spectrum data and the recent conversion-electron data. Stephens[10] reported gamma rays of 187 kev (0.5%) and 150 kev (0.8%). He also observed K x-rays, presumably due to the coversion of the two gamma rays, in such an abundance that at least one and probably both of the gamma rays are magnetic in nature. The recent conversion-electron study by Stephens and Asaro[70] showed the presence of gamma radiations of 36.60, 38.53, 62.97, and 99.53 kev. The gamma rays fall well into place in the level scheme described by the alpha-particle work, with but one exception. The existence of two low-energy transitions indicated by the conversion-electron work is somewhat troublesome. Possibly this indicates that there are two close-lying levels at about 37 kev. As these levels would be very close together, the alpha-spectrum measurements would not have given any indication of their existence. As there is no other evidence for two close-lying levels, only the 36.60-kev transition is placed in the decay scheme. The tentative level indicated

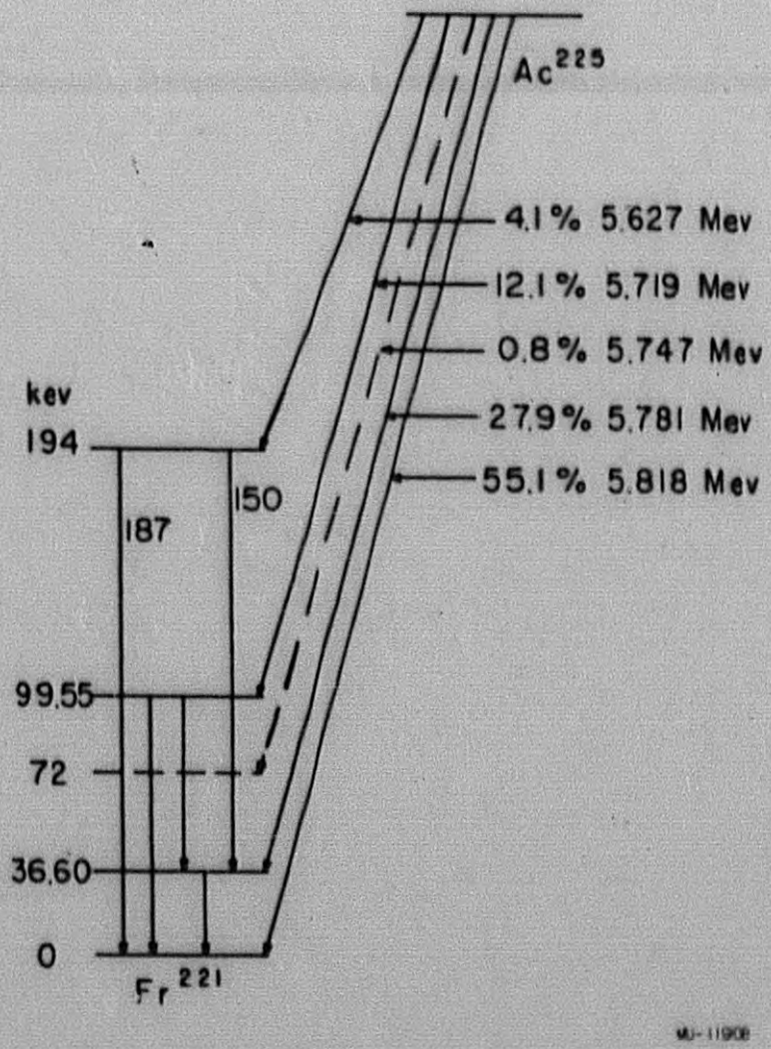

Fig. 38. Decay scheme of Ac^{225}.

in the new alpha-particle studies is noted in Fig. 38. There is no evidence for any mode of deexcitation of this state. However, because of the small alpha population to that state, it would be difficult to observe any of the possible transitions.

The conversion-electron data allow one to make multipole order assignments for some of the gamma transitions. The 36.60- and 38.53-kev transitions appear to be E2 in nature. The 62.97-kev transition is probably M1. Thus, the predominance of one type of parity is clearly indicated. It is not possible to make spin assignments for any of the levels in Fr^{221} at this time.

I. Alpha Decay of Thorium-227

The study of the alpha-particle fine structure associated with the decay of Th^{227} is still in progress. However, the preliminary data from this study are presented mainly to show deviations from previous work on this isotope and are later to be discussed in reference to alpha decay theory.

Rosenblum and associates[72] have made a study of the alpha-particle spectrum of Th^{227}, resulting in the assignment of eleven alpha groups to this decay process. This represents one of the most complex alpha spectra known. These eleven alpha groups are distributed over an energy range of 380 kev. Their intensities range from 21% to ~1%. Table XXXII summarizes these data, showing the absolute energy, the decay energy to the highest-energy transition (the assumed α_0), and the abundance of each group.

The Th^{227} used in this study was milked from its 22-year parent Ac^{227} and subsequently purified from its daughter activities. This purification was done by using a combination cation column and solvent-extraction technique. After separation, the Th^{227} was vaporized in the usual manner, with special care taken to get as thin a sample as possible (low vaporizer geometry). To date, three samples have been prepared this way.

Table XXXII

Alpha spectrum of Th^{227} (Rosenblum et al)

Energy (Mev)	Decay energy to a_0 (kev)	Abundance (%)
6.030	0	19
6.001	30	5
5.972	59	21
5.952	79	13
5.922	110	~2
5.860	173	4
5.796	238	2
5.749	286	17
5.728	307	~1
5.704	332	15
5.651	386	~2

The main problem here is to view the Th^{227} spectrum with as little interference as possible from Ra^{223}, which has alpha groups overlapping the lower-energy groups of the Th^{227}. Thus in each case, a short exposure of 1-hour duration was made as soon as possible (about 3 hours) after the chemistry to view the spectrum essentially free of Ra^{223}. Later exposures were made to observe the growth of the daughter alpha groups. To date, only three exposures have been evaluated.

Two of the three runs that have been scanned were made with the same sample (5×10^8 d/m). A high-resolution arrangement was used, giving peak half widths of 7 or 8 kev (18-mil defining slit). The first exposure was a 1-hour run made 3 hours after the separation, and the second exposure was a 10-hour run started 5 hours after the completion of the separation. The spectra from both of these exposures

are shown in Fig. 39. The dashed line outlines the spectrum obtained with the 10-hour run, and the solid line shows the spectrum from the 1-hour-long exposure. These spectra, with a few exceptions, are very similar to Rosenblum's. The 13% group reported by Rosenblum at 5.952 Mev is present (group Number 4), but it is in a much lower intensity than 13%. The group reported to be at 5.728 Mev in ~1% abundance appeared in the first run, but only in 0.1% intensity. Ra^{223} is known to have a 9% group in this region (5.730 Mev).[72] In the first run the Ra^{223} should be grown in to an extent of about 1% of the Th^{227} activity. The entire group at 5.728 Mev could be attributed to Ra^{223}, and this is the interpretation that has been made. The fact that the intensity of this group is lower than Rosenblum's estimate by a factor of 10 puts added faith in this interpretation. Also, on the short run, the peak at ~5.705 Mev shows definite indications of being complex. Ra^{223} has an alpha group in this region (5.704 Mev, 53%), but its expected intensity would be too low by a factor of 5 or 6 to explain the complexity of this peak. From these data, this peak is considered to be two Th^{227} alpha groups (Groups 10 and 11); there is no real evidence, however, that only two groups exist in this region. The longer exposure showed the group at 5.796 Mev to be complex also (Groups 7 and 8). Since Ra^{223} is not known to have an alpha group in this region, the new group has been assigned to Th^{227}. Also, in the long exposure, a rather broad peak is seen at the low-energy edge of the spectrum. Ra^{223} is known to have a 24% group in this region (5.596 Mev). The Ra^{223} contribution during this exposure would have been about 5.5%, sufficient to account for most if not all of that peak. However, the distinct broadness of the peak leads one to suspect that a low-intensity Th^{227} component may also be there.

The third exposure was made with a sample having 7×10^9 disintegrations per minute. In order to improve the resolution on this run, a 5-mil defining slit was used, and the baffle opening (see Fig. 1) was decreased from 2.5 inches to 0.5 inch. This decreased the effective transmission of the spectrograph by a factor of 10, so a more active sample was needed. The exposure was 1 hr in duration and was run

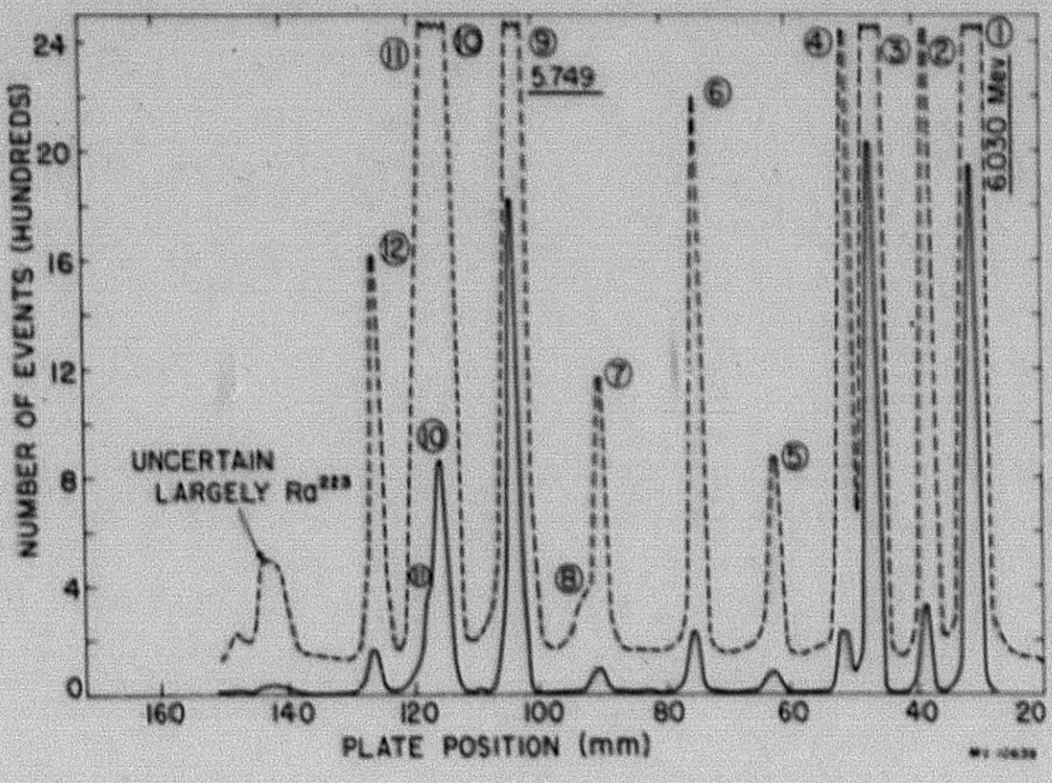

Fig. 39. Th^{227} alpha-particle spectra at high resolution.

about 3 hr after the chemical purification. The spectrum obtained in this case looked very similar to the spectrum gotten from the 1-hr exposure shown in Fig. 39, with two exceptions. First, the peak half widths were far superior (2.3 kev versus 7 kev). Secondly, the complex group labeled 10 and 11 in Fig. 39 was resolved into four components instead of two. A close comparison between this peak in the 1-hr exposure in Fig. 39 and in the higher-resolution run is shown in Fig. 40. Again, the Ra^{223} contribution to this region is small compared with the intensities of the four Th^{227} groups.

This preliminary work has led to some revisions in the complex alpha spectrum of Th^{227}. Table XXXIII summarizes the data from these three runs. There, the absolute energy, the decay-energy separation from the assumed a_0, and the abundance are given for each of the fourteen alpha groups. (This table can be readily compared with Table XXXII, which shows Rosenblum's results). As no attempt has been made in this study to determine the absolute energies of the Th^{227} alpha groups, the highest-energy alpha group reported by Rosenblum (6.030 Mev) was used as an energy standard in making up Table XXXIII.

As mentioned earlier, the extreme complexity of this decay process makes it difficult to present a decay scheme at the present time. One thing evident already from the conversion-electron study by Pilger[73] is that more levels than seen in the alpha-particle spectrum work may need to be postulated to account for the observed transitions.

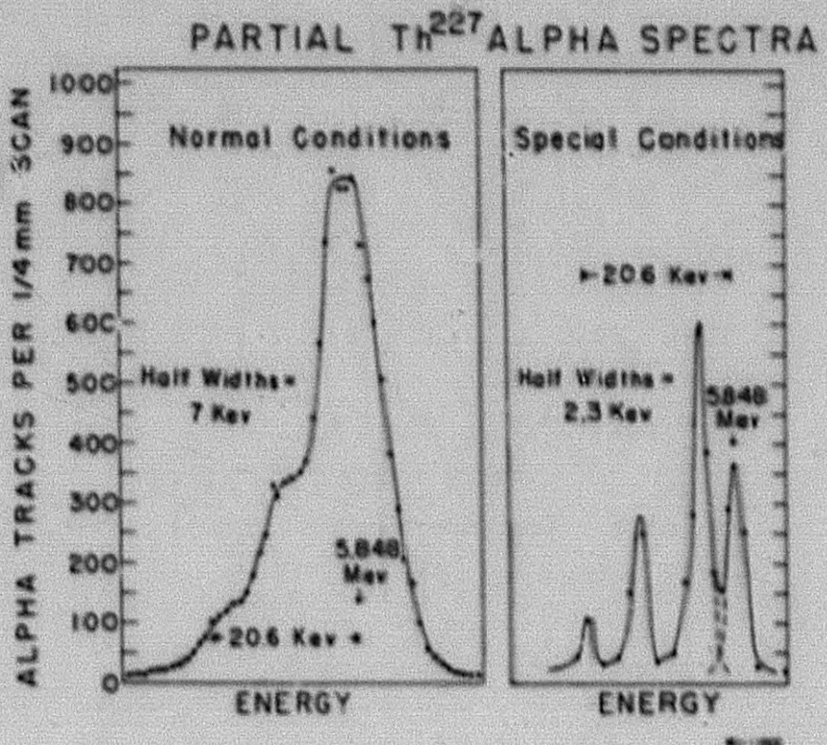

Fig. 40. Partial Th^{227} alpha spectra comparing the complex group under different resolution conditions.

Table XXXIII

Complex alpha spectrum of Th^{227}

Energy (Mev)	Decay energy to α_0 (kev)	Abundance (%)
6.030	0	24
6.001	30	4
5.970	61	25
5.951	80	3
5.908	124	1
5.859	174	3
5.800	234	1
5.792	242	0.1
5.749	286	23
5.706	330	4
5.702	334	6
5.693	343	3
5.686	350	1
5.661	376	2

J. Miscellaneous Studies

1. Alpha Decay of Polonium-206

Po^{206} has a 9-day half life[74] and decays predominantly by electron capture to Bi^{206}. A rough determination of the alpha branching ratio gave about 10% of the decays[74] going by alpha emission to Pb^{202}. Rosenblum[75] studied the alpha decay of Po^{206} with his alpha-particle spectrograph and reported two alpha groups, 5.218 Mev (96%) and 5.064 Mev. (4%). This would put the first excited state at 156 kev in the Pb^{202} nucleus.

A plot of the energies of the first excited states of even-even nuclei is given in Fig. 41. This plot is one prepared by F. Asaro in 1953, and contains the data up to that time. One notes the very rapid fall-off in the energies of the first excited states of the even-even lead isotopes with decreasing neutron number below 126 neutrons. A re-examination[76] of the Pb^{204m} decay scheme has since showed that the first excited state in this nucleus is at 890 kev rather than the previously reported 374 kev. This, of course, cast some doubt on the reported 156-kev first excited state in Pb^{202}. It was therefore deemed necessary to reinvestigate the alpha-particle spectrum of Po^{206}.

The Po^{206} for this series of experiments was obtained by milking it from Em^{210}. The Em^{210} was made by the spallation of thorium metal by 340-Mev protons in the Berkeley 184-inch cyclotron. The emanation fraction was isolated by freezing it out in a vacuum line. The emanation collected in this way was predominantly $Em^{208, 209, 210}$, and Em^{212}. The Em^{210} has a 2.7-hour half life and decays predominantly by alpha emission to Po^{206}. The other emanation isotopes have half lives of about one-half hour.[43] Thus, if the emanation is allowed to decay for 3 or 4 hours, it is essentially all Em^{210}. After allowance for the decay, the emanation is purified and the Po^{206} allowed to grow in. The Po^{206} made in this way can be gotten essentially free of other polonium isotopes. The polonium was then treated in two different ways to prepare sources for the alpha-particle spectrograph. Two sources were made by plating the Po^{206} onto a 10-mil silver wire by

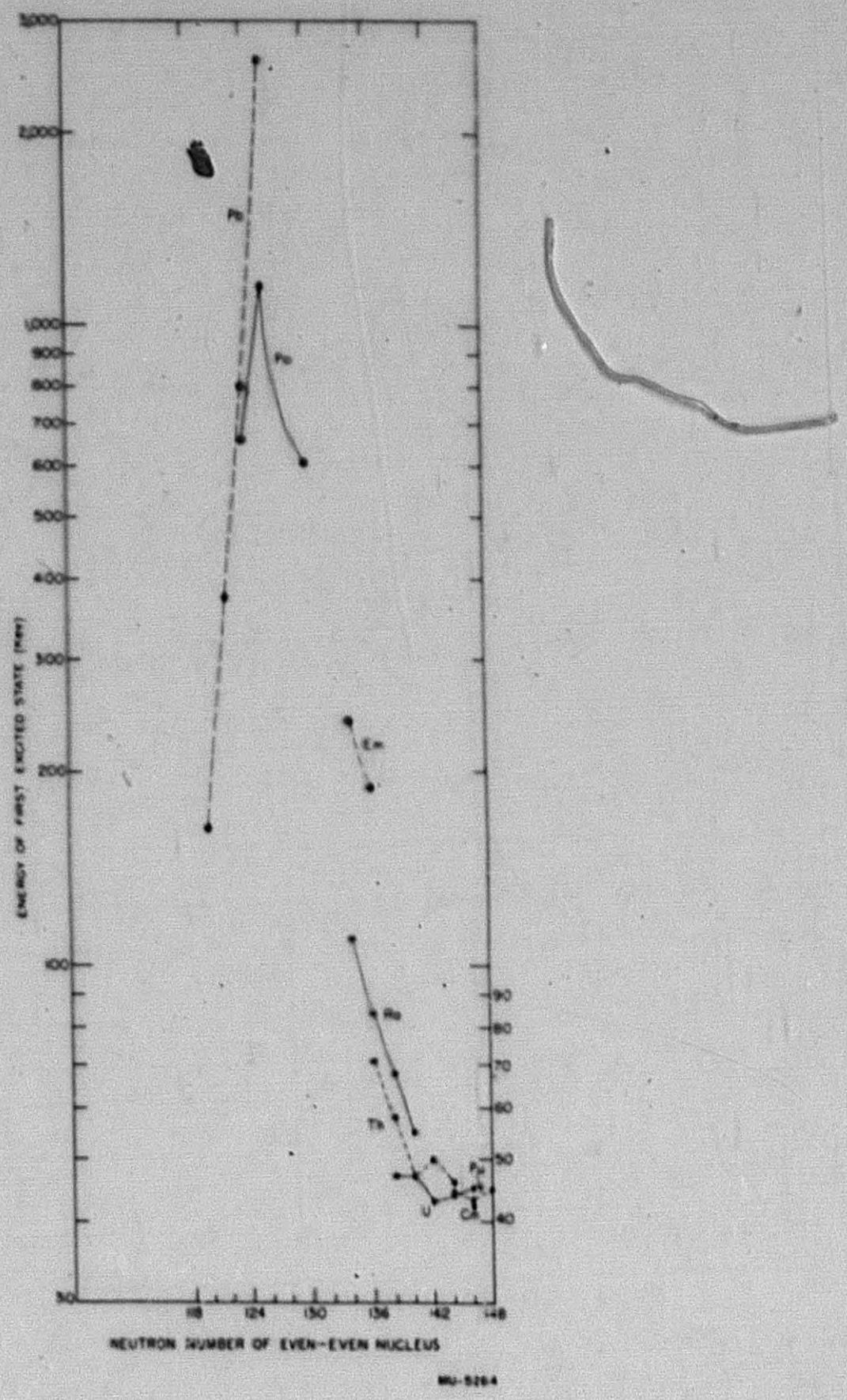

Fig. 41. Energies of first excited states in even-even nuclei (Asaro, 1953).

soaking the wire in a slightly acidic solution of the polonium. Two other sources were prepared by the vacuum-sublimation technique.

The first sample was on a silver wire and contained 7.5×10^5 alpha disintegrations per minute. The sample was exposed for 2 days in the alpha-particle spectrograph. Although there was a large amount of low-energy tailing due to sample thickness, there was no evidence for the previously reported fine structure. Only one alpha group was seen, corresponding to the 5.218-Mev ground-state transition seen by Rosenblum. Owing to the tailing, a limit of 3% on the presence of other groups in the region of interest (100 to 200 kev lower in energy than α_0) was set.

The second wire sample gave no information, because it contained only a small amount of activity.

The first vaporized sample contained about 10^6 alpha disintegrations per minute. As the emanation was not allowed to decay very long before purification, it was suspected that some Po^{208} would also be present in the sample. Po^{208} has an alpha group at 5.108 Mev,[77] which, if present to a large extent compared with the Po^{206}, might interfere in the search for the previously reported 5.064-Mev group of Po^{206}. The first exposure of this sample for 2 days did show the main Po^{206} peak and a group suspected of being Po^{208}. Even though the Po^{208} was present, no alpha group was seen at 5.064 Mev in abundance greater than 1%. A second 9-day exposure with this sample confirmed the assignment of the second group to Po^{208}, and further allowed a limit of 0.5% to be set on a second Po^{206} group at 5.064 Mev.

The second volatilized sample contained 5×10^5 alpha disintegrations per minute and contained no appreciable amount of Po^{208}. A 10-day run with this sample set a limit of 1% on the presence of a second alpha group for Po^{206} (Fig. 42). This limit holds to an energy 480 kev lower than the main Po^{206} group (5.218 Mev).

Stephens,[78] doing alpha particle-gamma ray coincidences on Po^{206}, sets a limit of 1.7×10^{-4} photons per alpha particle for the presence of gamma radiation in the 100- to 300-kev range. Thus, it seems pretty certain from the alpha data and coincidence data just

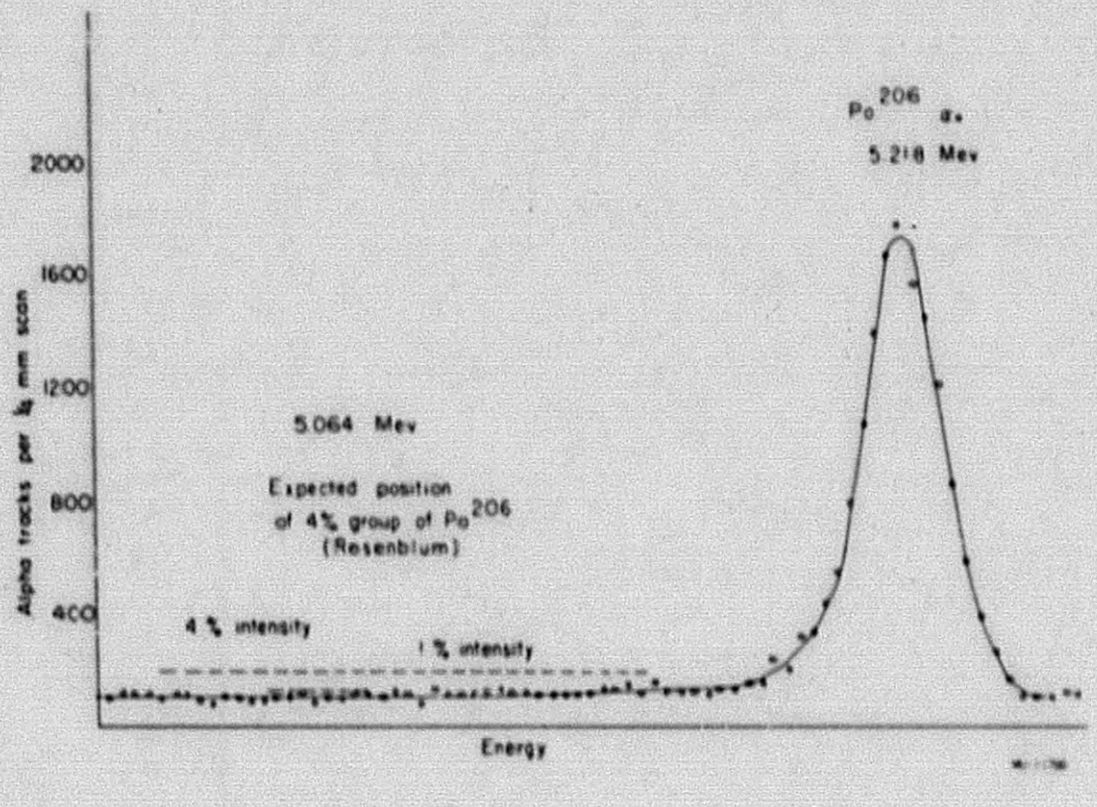

Fig. 42. Po^{206} alpha-particle spectrum.

presented that the previously reported 4% group in Po^{206} alpha decay does not exist, and that the first excited state in Pb^{202} is not at 156 kev as previously thought. Rosenblum[79] has stated in a later publication that he has not been able to confirm the presence of the second alpha group. However, he has not set a limit on its intensity.

The sample containing both Po^{208} and Po^{206} allowed a convenient check of the absolute energy of the main Po^{206} group. With the 5.108-Mev Po^{208} group as a standard, the two runs gave 5.216 and 5.219 Mev for the Po^{206} energy, in good agreement with Rosenblum's value.

2. Alpha decay of Astatine-209

At^{209} is a predominantly electron-capture nuclide with a half life of 5.5 hours.[80] Barton et al. estimated the alpha branching ratio to be about 0.05. A conversion-electron study by Mihelich et al.[81] yielded several gamma transitions, which were assigned to At^{209} on the basis of their 5.5-hour half life. A study of the alpha-decay fine structure of this isotope is of interest in order to determine whether or not all the reported gamma rays should be assigned to the electron-capture branch of the decay.

Samples of At^{209} were made by bombarding bismuth metal with helium ions in the Crocker Laboratory 60-inch cyclotron. The first sample was made by a bombardment with 38-Mev helium ions, and the second by 46-Mev helium ions. In the first case, the astatine fraction was vacuum-sublimed onto a platinum plate in the usual manner. The second sample was prepared by soaking a 10-mil silver wire in a slightly acidic solution of the astatine. Both sources were exposed in the alpha-particle spectrograph.

The first sample was exposed for about 13 hours and then again for 22 hours a day later. The first exposure showed two peaks, one at 5.86 Mev (500 tracks high) and one at about 5.64 Mev (50 tracks high). The peak at 5.86 Mev was presumably due to At^{211}, which was expected to be in the sample; this peak was then used as an energy standard (5.862 Mev)[82]. The other group was assigned to At^{209} because of its nearness to the ion-chamber-measured energy for At^{209} of 5.65 Mev

(Barton, Ghiorso, and Perlman). With the At^{211} group as an energy standard, the energy of the At^{209} group was 5.642 Mev. There was no evidence of any fine structure in the At^{209} alpha decay, but owing to the small amount of At^{209} activity an abundance limit of only 20% or 30% of the main group could be set, down to 5.25 Mev. Because of the rapid decay of the At^{209}, the second exposure did not show any alpha group at 5.64 Mev.

The second source contained about 300 times as much At^{209} as the first source, so that a peak height of 5600 tracks was observed with an 8-hour exposure. The sample was then exposed for 14 hours right after the first exposure ended. The At^{209} peak was 2000 tracks high on the second exposure, consistent with the known half life for this nuclide. Since At^{210} was also produced in this bombardment, the three At^{210} groups at 5.519, 5.437, and 5.355 Mev[82] (each peak about one-tenth of the At^{209} peak in height) and the Po^{210} group at 5.299 Mev[83] (comparable to the At^{209} peak in height) were also present. These groups were used as internal energy standards, giving the At^{209} group an energy of 5.642 Mev on each exposure. This energy determination should be good to about 5 kev, allowing for the uncertainties in the energies of the standards and uncertainties in locating the peaks on the photographic plate. The spectrum observed on the second exposure with this sample is shown in Fig. 43.

The presence of the At^{210} and Po^{210} groups somewhat obscures the search for alpha fine structure in the At^{209} decay. However, a limit of 2.5% abundance can be set, down to an energy 660 kev below the main At^{209} group. This limit is not valid in the immediate vicinities of the At^{210} and Po^{210} groups, i.e., 5.53 to 5.50, 5.45 to 5.42, and 5.37 to 5.26 Mev. It does seem highly probable, however, that no low-lying fine structure in large abundance exists in the At^{209} alpha decay, and that the observed gamma rays probably all belong to the electron-capture branch.

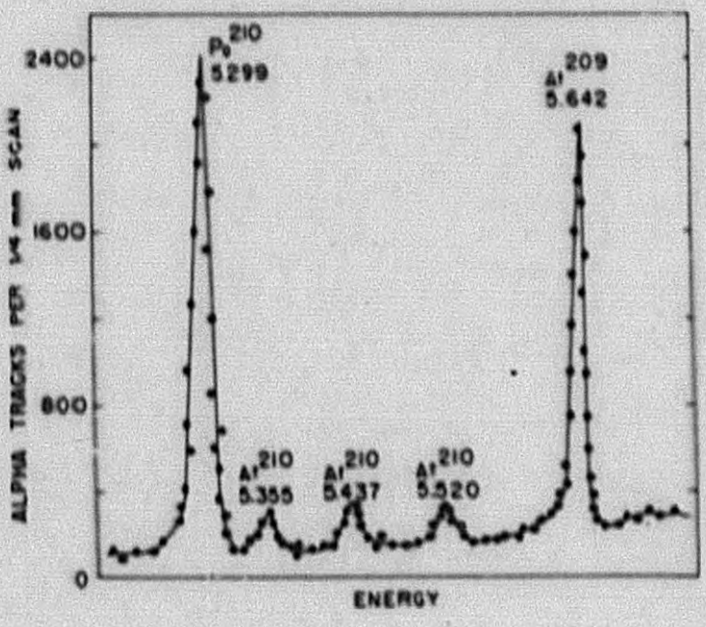

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Fig. 43. Alpha-particle spectrum of $At^{209,210}$ sample.

3. Alpha Decay of Thorium-230

Ionium has been the subject of a considerable amount of experimental interest for some time now. Being one of the natural radioactive species, it is readily available in large amounts, and this may have had some bearing on the interest shown in it. Rosenblum et al.[84] in France have studied the alpha spectrum of Th^{230} and report eight alpha transitions populating levels up to 485 kev above the Ra^{226} ground state. Recently Stephens[10] has undertaken a thorough study of the gamma radiations accompanying this decay, and reports some difficulty in interpreting his data in terms of the alpha spectrum reported by Rosenblum's group. Since large amounts of this nuclide were available, it was decided to attempt a reinvestigation of the complex alpha spectrum of ionium.

One of the main difficulties with ionium is its long half life (80,000 years[43]) and corresponding low specific activity (4.3×10^4 alpha disintegrations per minute per microgram). Any alpha components of several percent abundance could be fairly easily observed with samples of 10 μg of material or less in the alpha-particle spectrograph. But, for reasonable exposure times, weak components present to the extent of a few tenths of a percent would be observed only with very large amounts of material (about 100 μg). Several samples of less than 10 μg each were vaporized, to be used in a study of the main groups. Several other large samples containing 50 to 100 μg of ionium were vaporized, to be used to study weak members of the alpha fine structure. The large samples were each exposed for 1 or 2 days in the alpha-particle spectrograph and the plates scanned to compare the low-energy tailing effects for each sample. The tailing effect was an important consideration in choosing a sample to be used for a long exposure, since the low-intensity groups in question are all in an abundance of 0.2% or less and are all lower in energy than the two intense groups. The sample with the least amount of tailing was chosen for some longer runs. It was expected that alpha groups of 0.1% abundance would be just observable with this sample.

The data taken from the smaller samples designed to show the main components of the spectrum are given in Table XXXIV (see Fig. 44). It is seen that two alpha groups in approximate abundance ratio of 3 to 1 account for the main part of the spectrum (as expected from Rosenblum's work). After one corrects the energy separation given in Table XXXIV for the recoil energy, a decay-energy separation of 67.8 kev is obtained. This is in good agreement with the French electron-study value of 67.76 kev[85] for the gamma transition from the first excited state to ground. From the intensity ratio given in Table XXXIV the highest-energy group accounts for 74% of the decay, and the group 67 kev away, 26%. Rosenblum reports 76.3% and 23.4%, slightly different from these values, but in good agreement with them.

The sample exposed in the longer runs to search for low-intensity groups contained 3×10^6 alpha disintegrations per minute. Two exposures, of 11 and 12 days, were made. No alpha groups other than the main groups just discussed were seen over the same energy range in which Rosenblum reports six low-intensity groups. However, in both cases, a limit of 0.2% on the maximum abundance of groups in this range is the best that could be set. All the low-intensity groups reported by Rosenblum are either 0.2% or lower in abundance, so that this work does not contradict that by the French group. The inability to set a lower limit on the abundances in this region is due to variations in the background, which were larger than anticipated. The background in this case was due about equally to tailing effects and to machine background.

For a decay scheme of Th^{230} the reader is referred to a recent article by Stephens, Asaro, and Perlman.[86]

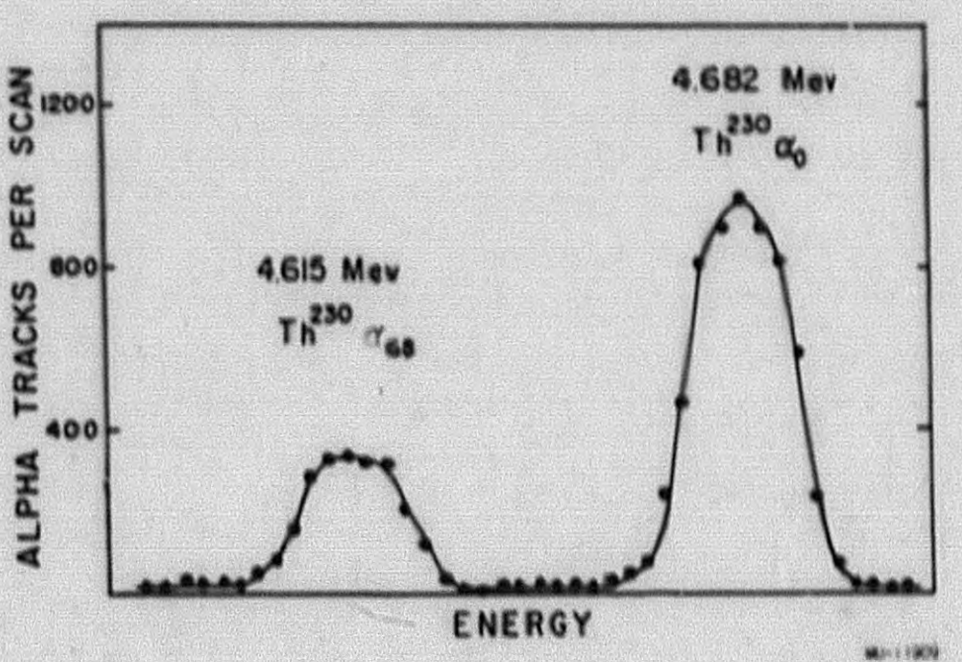

Fig. 44. Main alpha groups of Th^{230}.

Table XXXIV

Main alpha groups of Th^{230}

Exposure No.	Particle-energy separation (kev)	a_0/a_{68} abundance ratio
395	67.2	2.88
403	66.2	2.86
404	66.5	2.76
Best values	66.6 kev	2.85

4. Alpha Decay of Plutonium-242

Pu^{242} is a long-lived plutonium isotope which decays by alpha-particle emission. A wide range of half-life values have been reported for this isotope, ranging from 3.73×10^5 years[87] to 9×10^5 years.[88] The first estimate of this half life was 5×10^5 years,[89] which was determined from the yield of Pu^{242} milked from Am^{242m}. Another value of 3.78×10^5 years[90] has also been reported. Asaro's work[88] consisted of determining the activity ratios of the various plutonium isotopes in a mass-analyzed sample. This was done with the aid of the alpha-particle spectrograph. Thus, he also obtained information on the alpha-decay fine structure of Pu^{242}, reporting two alpha groups at 4.898 Mev (80%) and 4.854 Mev (20%). Since plutonium samples containing much larger amounts of Pu^{242} than used in the earlier study were readily available, a reinvestigation of this isotope was undertaken.

The Pu^{242} was made by intensive neutron irradiation of Pu^{239} in the Materials Testing Reactor at Arco, Idaho. This results in a mixture of plutonium isotopes whose isotopic consitution depends on the length of irradiation. The separated plutonium fraction was mass-analyzed[91] and shown to be mostly Pu^{242} (masswise). The results

of the mass analysis are given in Table XXXV. Two volatilized sources (each of about 10 μg of material) were prepared for exposure in the alpha-particle spectrograph.

Both sources were exposed in the alpha-particle spectrograph with the maximum-geometry slit arrangement. The first sample was exposed for 8 days, and the resulting spectrum is shown in Fig. 45. As expected from the mass analysis and relative half lives, both Pu^{242} and Pu^{240} showed up in comparable intensities. Two alpha groups appeared for each nuclide. Using the mass-analysis data, the relative activities determined from the alpha-particle spectrograph run, and the known half life for Pu^{240} (6.58×10^3 years),[92] one can calculate a half life for Pu^{242} by

$$T\,\frac{1}{2}\,(242) = T\,\frac{1}{2}\,(240)\,\frac{N_{242}}{N_{240}}\,\frac{A_{240}}{A_{242}}\ ,$$

where N_{242}/N_{240} is the mass ratio of the two isotopes and A_{240}/A_{242} is their activity ratio. In this way, a half life of 3.6×10^5 years was calculated for Pu^{242}. From a 5-day run on the second source, a value of 3.4×10^5 years was obtained. The first value is probably best, considering that about twice as many alpha tracks were recorded in that run as in the second. It should be mentioned that corrections were made in these calculations for the difference in spectrograph geometry with position on the photographic plate (see the dicussion of the Am^{243} half-life determination). Also, corrections were made for the small contributions of Pu^{239} to the Pu^{240} peaks and Pu^{241} to the Pu^{242} peaks. These were made by using the mass-analysis data and the known half lives for the isotopes. The corrections for both Pu^{239} and Pu^{241} were about 1% of the Pu^{240} peak. These half lives just determined are in good agreement with the lower values of the previous measurements mentioned earlier. That the previous spectrograph determination is in poor agreement with these values may be laid to the unfavorable ratio of Pu^{242} to the other plutonium isotopes (activitywise) in that sample.

The spectrum shown in Fig. 45 showed the better resolution of the Pu^{242} fine-structure components. In this run the two alpha groups

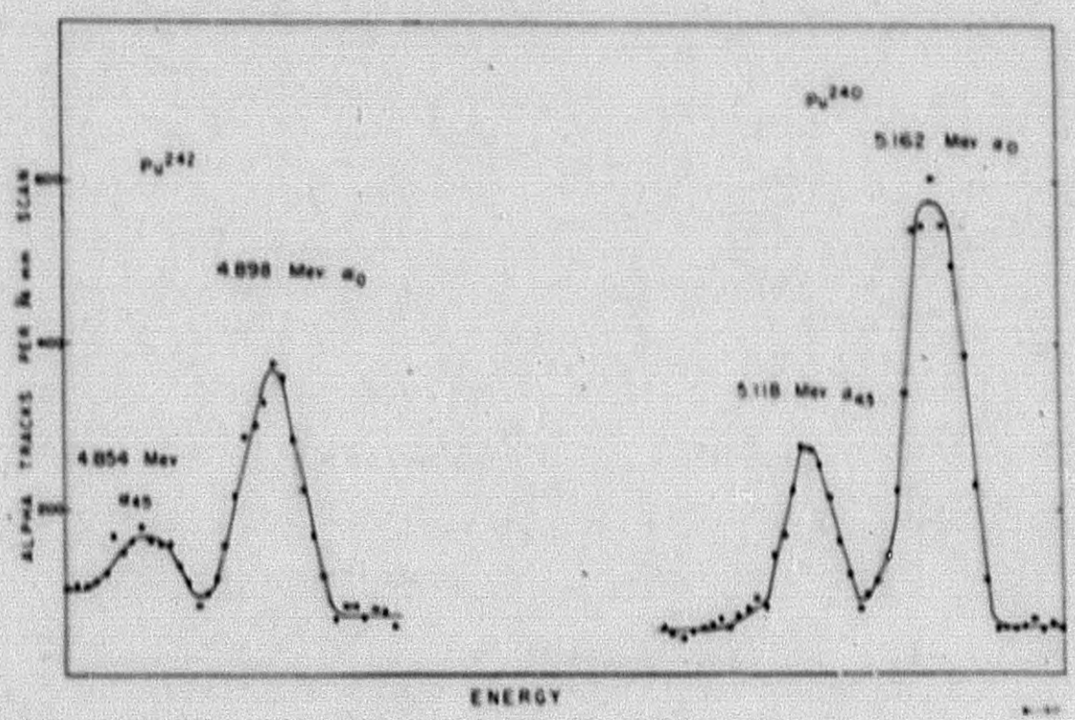

Fig. 45. A portion of the alpha-particle spectrum of a sample of mixed plutonium isotopes.

were separated by 44 kev, in good agreement with Asaro's separation.[8] The abundances of the two groups were 74% for the highest-energy group and 26% for the other group. Asaro had previously reported 80% and 20% abundances. The new values are probably better than those of Asaro, considering the larger amount of Pu^{242} activity used in these runs. The assignment of the alpha group at 4.854 Mev to Pu^{242} was not considered definite at the time of Asaro's work. This assignment is now confirmed. The higher-energy group is probably the ground-state transition and the other group probably populates the 2+ level of the ground rotational band. It should be pointed out that one would not expect to observe alpha transitions to higher states with the present samples, since such transitions would be expected to be in very low intensity.

Table XXXV

Mass analysis of plutonium sat.

Isotope	Abundance (%)
238	0.258
239	0.126
240	3.15
241	1.96
242	94.49
244	0.0232

IV. DISCUSSION OF RESULTS

There have been a number of regularities apparent in the decay properties of the heavy even-even alpha emitters. The regularities concern not only the nature of the energy levels populated, but also the alpha-transition intensities to the various levels. The odd-mass nuclei in this region have not shown regular behavior as a general rule, although some features of their decay properties are similar. The two classes of isotopes are discussed separately.

A. Even-Even Nuclei

One of the striking features in the region from emanation to the higher elements is the occurrence of a level structure best explained in terms of the collective model of the nucleus.[42] The very low energies of the first excited states in this region have been considered as evidence for collective phenomena. Beyond this, the properties of other low-lying excited levels also fit in well with this picture. From the collective approach, groups of levels belonging to a rotational band and having an I(I + 1) energy dependence are expected in this region. For the ground-state band, the expected levels would be 0+, 2+, 4+, etc. These levels would be expected to de-excite by E2 cascades. The even-even isotopes studied in this research are considered in the light of the collective approach.

Considering the I(I +1) energy dependence for a moment, one would expect the ratio of the energy of the 4+ state to that of the 2+ state to be 3.33. Perlman and Asaro[6] have plotted this ratio as a function of neutron number, and the 3.33 value seems to be obeyed in nuclei above radium. The data taken for U^{232} (from Pu^{236} alpha decay), Pu^{240} (from Cm^{244} alpha decay), and Cm^{242} (from Cf^{246} alpha decay) in this study also show the 3.33 ratio for the energies of the first two excited states. One also notes that the gamma transitions from these levels are consistent with the expected behavior for members of rotational bands. In two cases, tentative assignments of a third excited member of a rotational band have been made on the basis of observed gamma rays. If these are the 6+ members, the ratio of their energies and the 2+ state energies should be 7.

The observed ratios are seen to be very close to 7. In most cases involving rotational bands in the heavy-element region, no rigorous experimental proof that the spins are indeed 0, 2, 4, etc. has been obtained, and the assignment of the spins to these levels has been based mostly on their observed energy ratios and gamma-ray de-excitation patterns.

Another interesting point in connection with the even-even alpha emitters is the status of alpha-decay theory. The one-body model for alpha decay proposed by Gamov[93] and by Condon and Gurney[94] considers alpha decay as a barrier-penetration problem. This treatment introduces a penetration term, as the most important factor, that is dependent on the energy of the alpha particle, the nuclear radius, and the nuclear charge. The equations resulting from this approach have been put into several different forms, and they are not discussed here in any detail. However, they make it possible to predict the half life that an alpha transition of a given energy should have if the radius of the particular nucleus is known. Since nuclear radii are not known in general in the heavy-element region, the observed alpha-decay half lives and energies have been used to calculate the nuclear radii. Generally the ground state-to-ground state transition is used in these calculations. If one uses the nuclear radius calculated in this manner and calculates expected half lives for other transitions observed in the same decay, systematic deviations from the experimentally observed half lives are noted. The deviations are generally expressed in terms of hindrance or departure factors, which are the ratios of the observed and calculated partial alpha half lives.

A plot of these hindrance factors is given in Fig. 46, wherein separate lines are drawn for the transitions to the 2+ levels and to the 4+ levels. The plot in Fig. 46 is made against mass number, rather than proton number or neutron number as previously done.[6, 95] A fairly smooth trend for increase in the hindrance factors with increasing A is noted in both cases. One quickly notes that the maximum in the 4+ curve is reached for Cm^{244} decay (this research). As yet, no maximum has been reached in the 2+ hindrance factors. The significance of such curves has not as yet become evident. Rasmussen[96] has had a

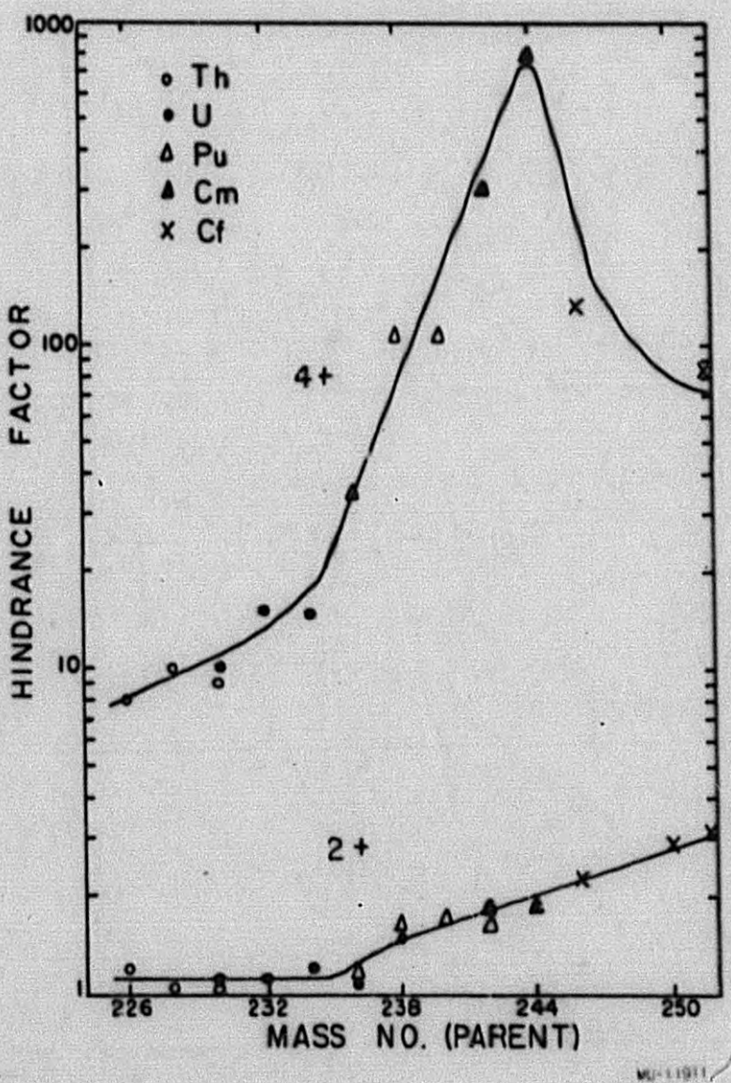

Fig. 46. Hindrance factors for alpha transitions to the 2+ and 4+ levels in even-even nuclei.

certain amount of qualitative success in considering the interaction between the outgoing alpha-particle wave and the intrinsic nuclear quadrupole moment. He finds that the alpha population to the 4+ state should be depressed, depending upon the amount of quadrupole distortion. Such coupling should cause the alpha-particle waves of various angular momenta to go through nodes, which may explain the maximum seen in the 4+ curve of hindrance factors.

The data obtained in this study for the heavy even-even alpha emitters seems to be very consistent with the well-established regularities characteristic of the group. The validity of the collective model for this class of nuclei has also been reaffirmed.

B. Odd-Mass Nuclei

As a general rule, odd-mass nuclei are not yet as well systematized as the even-even nuclei. The collective model also predicts rotational bands for odd-mass nuclei, in which case the energy spacing is again $I(I + 1)$. In the odd-mass case the allowed spin values are I_0, $I_0 + 1$, $I_0 + 2$, etc. (all of the same parity), where I_0 is the intrinsic spin of the base level of the band. A well-characterized case showing rotational structure in this region is that of Np^{237} (alpha daughter of Am^{241}). The similarity between Np^{237} levels and those in Np^{239} has already been pointed out. Rotational structure is also evident in Pu^{239},[97] Th^{229},[6] and Bk^{249}. One notes a wide difference in the patterns of the alpha decay to these nuclides. In Np^{237} and Np^{239}, the alpha decay populates most prominently a rotational band that is not the ground-state band. On the other hand, U^{233} and E^{253} decay seem to populate ground-state rotational bands.

Also of interest for odd-mass nuclei is the status of alpha-decay theory. In most cases, it is found that the ground-state alpha transitions are hindered. Perlman and Asaro[6] have assembled the hindrance factors for these ground-state transitions, and they vary greatly in magnitude. There seems to be no systematic way in which they vary from one nuclide to another. In most cases an unhindered alpha transition is seen to at least one excited state. In the decay of Am^{243}, the unhindered decay

populates the 74-kev level (the ground-state transition is hindered by a factor of 1600). In Am^{241} decay the unhindered alpha transition populates the 60-kev level of Np^{237}. In Pa^{231} decay unhindered decay is observed to the 329-kev state of Ac^{227}. E^{253} decay is quite different in that the unhindered alpha decay populates the ground state of Bk^{249}. Another quite different situation is observed in the Th^{227} alpha decay, where--in spite of all the levels apparently available for alpha population--no unhindered transition has as yet been observed. Bohr, Froman, and Mottelson[98] have considered the unhindered alpha transitions in odd-mass nuclei to be $\Delta I = 0$ transitions involving the same nuclear states in the parent and daughter nuclei.

Where the favored alpha transition is decaying from a parent in state I_i, K_i to a daughter state I_f, $K_f = K_i$, Bohr et al.[98] give the following relationship for the transition probability:

$$P = P_0(E) \sum_{\ell} c_\ell \langle I_i \ell K_i 0 / I_i \ell I_f K_f \rangle^2 .$$

The quantity $P_0(E)$ is an energy-dependence factor (from simple alpha-decay theory). The c_ℓ's are constants that determine the total reduced transition probability for a given ℓ. They can be determined from the observed transition probabilities in near-by even-even nuclei. The value of c_0 is taken as unity by the definition of P_0 ($\ell = 0$ alpha-particle transitions are assumed to be unhindered in even-even nuclei). Bohr et al. have made calculations of the expected intensities to members of the same rotational band, using the above relationship, and they report satisfactory agreement with the observed intensities for several cases. In doing these calculations, they have assumed a value of 0.7 for c_2. This corresponds to a hindrance factor of 1.4 for the $\ell = 2$ transitions, which correspond to the 2+ levels mentioned in the even-even nuclei discussion. The hindrance factors for the $\ell = 2$ transitions were seen to be appreciably larger than this for the higher elements.

There are two cases where extremely good data are available on the fine structure of alpha decay to rotational bands in odd-mass nuclei. These are the decays of Am^{243} and Am^{241}. Also, very good data are available on the near-by even-even isotopes. For these two cases, the calculations were redone. To determine the values of the c_l's to use in each case, an average of the observed $l = 2$ and $l = 4$ hindrance factors for the neighboring even-even nuclei was taken. This gave c_2's of 0.56 and 0.59, and c_4's of 0.002 and 0.005, for Am^{243} and Am^{241}, respectively. With these constants, the expected intensity ratio for alpha decay to the 5/2, 7/2, 9/2 members of the main rotational band in Am^{243} decay is 100/12.0/1.9. The observed ratio is 100/13.2/1.5. This agreement is within the experimental errors involved in determining the c_l's and in determining the Am^{243} abundances. For Am^{241}, one calculates an intensity ratio of 100/13/2.2, in which case the observed ratio is 100/16/1.7. The agreement here is not so good as for Am^{243}, but is probably within the experimental errors involved. If one considers U^{233} decay,[6] which populates a 5/2, 7/2, 9/2 rotational band in Th^{229}, using a c_2 of 0.87 and a c_4 of 0.067, an intensity ratio of 100/16/2.9 is obtained. This is to be compared to the experimentally observed ratio of 100/18/2.4, again showing very good agreement. This type of agreement, using the approach outlined by Bohr et al., makes one conclude that the unhindered decays observed in the odd-mass nuclei are indeed $\Delta I = 0$ transitions. Thus, the rotational bands receiving the majority of the alpha population in the odd-mass nuclei seem to represent nuclear states that are quite similar to the ground states of the parents.

It is somewhat interesting to speculate about the origin of the nuclear states observed in these decay studies. From the striking rotational features observed in this region, it is quite evident that we are dealing with deformed nuclei. Mottelson and Nilsson[99] have considered the effects of distortion upon the ordinary shell model, and have shown that the spherical shell-model levels split into several components in the distorted nucleus. These components are classified according to Ω, which is the component of the nucleon's angular momentum along the nuclear symmetry axis. A plot of the energies of these new states as a

function of prolate deformation shows that the high-Ω components from a given state increase in energy with an increase in deformation, while the low-Ω components from the same level decrease in energy. For large deformations, the $\Omega = 11/2$ component of the $h_{11/2}$ state (below the 82-nucleon closed shell) overlaps in energy with levels arising from single-particle states above the closed shell. In considering the possibility of an 11/2 spin for the Bk^{249} ground state indicated by the level spacings observed in the E^{253} alpha decay, one has to keep in mind the possible proton states above 82 protons. One could use the $\Omega = 11/2$ state arising from the $i_{13/2}$ single-particle state or the $\Omega = 11/2$ state from the $h_{11/2}$ state (discussed above). Since high-Ω components from the $i_{13/2}$ state rise rapidly with deformation, they are out of reach for 15 protons beyond the closed shell of 82. On the other hand, with reasonable distortions, the $h_{11/2}$ component could be available for filling at 97 protons. Thus, the presence of a high spin state fairly close to the 82-proton closed shell is easily explained in terms of the particle states available in a deformed potential. It should be stated that others have had a reasonable amount of success in assigning states observed in other nuclei in this region in terms of the Nilsson approach.[100]

The two extremely complex decay schemes seen in this work, Th^{227} and Pa^{231}, are rather interesting in that no prominent rotational features are seen in their spectra. Possibly interactions between states have distorted the rotational features (energy spacings and alpha-particle intensities) to the point where they are no longer recognizable.

With the exception of the extremely complicated cases, many of the features of the odd-mass alpha-decay schemes find their explanations in the collective model and associated theories. It will be interesting to see if further extensions of these theories can explain the features of the more complicated cases.

V. ACKNOWLEDGMENTS

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