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Frank Asaro, F. L. Reynolds, and I. Perlman
February 16, 1952

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#### ABSTRACT

The alpha particle spectra of  $Am^{241}$  and  $Cm^{242}$  was studied in detail utilizing a 75 cm radius of curvature 60° cymmetrical electromagnetic analyzer with photographic plate detection. The radioactive sources containing up to 3  $\mu$ g/cm<sup>2</sup> of active atoms in the case of Am<sup>2 $\mu$ 1</sup> were prepared by vacuum sublimation. The average geometry of the spectrograph is about 4 parts in 105. The energy dispersion on the photographic plate is about 3.4 kev/mm for 5 Mev alpha particles and the width at half maximum of these alpha particle groups is about 7 kev. Six alpha particle groups were found in Am<sup>241</sup>, and their energies and abundances are 5.546 Mev. 0.23%; 5.535, 0.34%; 5.503, 0.21%; 5.476, 84.2%; 5.433, 13.6%; 5.379, 1.42%. In Cm<sup>242</sup> two alpha particle groups were found whose energies and abundances are 6.110 Mev, 73%, and 6.064, 27%. The alpha decay scheme is correlated with gamma rays and conversion electrons observed in this laboratory for both Am<sup>241</sup> and Cm<sup>242</sup>. The various alpha groups are evaluated with respect to the alpha decay systematics, and the degrees of hindrance of the various alpha transitions are discussed with reference to normal trends in even-even nuclei. It is suggested that the totally different patterns of the spectra of the two nuclides are conditioned by the nuclear types.

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#### TNTRODUCTTON

According to simple alpha decay theory<sup>1,2</sup> the predominant factor governing the decay constant is the decay energy. On this basis one might expect that alpha transitions other than those to the ground state of the product nucleus would compete poorly with the most energetic alpha particle so that complex spectra would be difficult to discern particularly if the spacing between nuclear energy levels is wide. A study of the decay characteristics of the many alpha emitters now known has shown that the even-even nuclides are in remarkable agreement with the demands of the theory while other types are not. This is not to say that the even-even nuclides show no complex structure but where multiple alpha groups are encountered the partial decay constants have been found to be in general agreement with expectations according to the energy for each group. The experimental manifestation of this relationship is that the ground state transition is most abundant whereas a transition by a lower energy alpha group is found in lower abundance.<sup>3,4</sup>

<sup>&</sup>lt;sup>1</sup>G. Gamow, Z. Physik <u>51</u>, 204 (1928); G. Gamow and C. L. Critchfield, <u>Theory of Atomic Nucleus and Nuclear Energy-Sources</u>, (Oxford University Press, London, 1949).

<sup>&</sup>lt;sup>2</sup>E. U. Condon and R. W. Gurney, Phys. Rev. <u>33</u>, 127 (1929); Nature <u>122</u>, 439 (1928).

 $<sup>^{3}</sup>$ Perlman, Ghiorso, and Seaborg, Phys. Rev.  $\underline{77}$ , 26 (1950).

 $<sup>^4</sup>$ T. Perlman and T. J. Ypsilantis, Phys. Rev.  $\underline{79}$ , 30 (1950).

There is mounting evidence as reviewed in an earlier publication<sup>3</sup> that those nuclides which have one or more unpaired nucleons depart from the regularities of the even-even type. It was found to be the rule that the alpha decay of the odd nucleon types was hindered in the sense that the half-life of each such nuclide was longer than predicted by the simple theory and that actually found for an even-even alpha emitter of the same atomic number and decay energy. A further generalization was suggested that has to do with complex structure of nuclides with odd nucleons and that is that the ground state transition is most highly hindered whereas one or more of the lower energy groups are present in relatively higher abundance than would be expected according to their respective energies. In fact, cases were found in which the ground state transitions were rare and it could be inferred that there might be instances in which the ground state transitions have not yet been discerned. The relatively long partial half-life of the ground state transition will in itself make complex structure more prominent. In the absence of the means of resolving alpha groups of similar energies the presence of abundant gamma radiation was often the best evidence for complex alpha decay.

The reason advanced<sup>3</sup> for the marked differences between the different nuclear types lay in a provision for the assembly of the components of the alpha particle. It was suggested that the time for assembly in <a href="even-even">even-even</a> nuclides is either negligible or nearly constant and that the theory cannot distinguish between the two. If one or two odd nucleons are present, however, it was postulated that additional time is required for the assembly of the alpha particle for the ground state transition since for this transition the odd nucleon either must be a participant or the necessary nuclear

rearrangement must accompany the process in order to leave the product nucleus in its ground state.

In the absence of the means for placing this explanation upon a quantitative basis we can examine in greater detail the complex spectra of both types of alpha emitters with the hope that further correlations will appear which will point toward an understanding of the factors at play. For example, the odd nucleon alpha emitters may show particular energy spacings in their complex structure and degree of hindrance in emission which can be correlated in terms of assigned states of the nucleons.

It is for these objectives in furthering alpha decay theory, in the possibility of assigning quantum states to the nuclides as well as for the intrinsic interest in the decay properties of heavy nuclides, that a program has been undertaken to measure complex structure of alpha emitters. In so doing the classical work on alpha particle spectroscopy will be extended to the many artificially produced alpha emitters.

For the measurements, a magnetic spectrograph has been constructed the details of which have been described elsewhere. The most detailed measurements to date have been made on Am<sup>241</sup> and Cm<sup>242</sup> which are representatives of the <u>odd-even</u> and <u>even-even</u> nuclear types, respectively. The availability of these nuclides in sufficient quantity has dictated their early selection.

#### EXPERIMENTAL

#### The Alpha Particle Spectrograph

The instrument used in the present measurements employs a 60° sector electromagnet and the normal trajectory has a radius of curvature of 75 centimeters. Figure 1 is a schematic diagram of the optics of the instrument

<sup>&</sup>lt;sup>5</sup>F. L. Reynolds, Rev. Sci. Instruments <u>22</u>, 749 (1951).

and shows the magnet, source, slit system, and detector. A more detailed description of the instrument, its power supply and operating characteristics have been published elsewhere. A brief summary of the characteristics are given here: the magnet power supply is capable of maintaining the current constant to 1 part in 10,000 over at least a 24 hour period. The detection system employed consists of nuclear emulsion plates in which the alpha particle tracks are counted using 450 power magnification. The dispersion at a given point on the detection plate varies linearly with particle energy and amounts to about 3.4 kev per millimeter at 5 MeV. The width at half maximum for the alpha particle peaks depends strongly upon the sample preparation and to a lesser degree upon the slit and baffle systems employed. These factors will be discussed below. The geometry factor has been varied between 10<sup>-6</sup> and 10<sup>-4</sup> and depends upon the slit and baffle openings.

#### Sample Preparation

In order to take advantage of the inherent high resolution of an alpha particle spectrograph it is imperative that the sources be extremely thin. Poor samples with respect to self-absorption are manifested by a tail on the low energy side of the distribution curve whereas the form for a thin preparation approaches a symmetrical peak. Among the natural radioactivities good samples have been prepared when applicable by collecting the active deposits from the emanation in the decay series.

In general, poor samples are obtained when prepared from solutions by simple evaporation of the solvents. Even when the weight of the sample corresponds only to the order of a few micrograms per square centimeter on the plate, the formation of micro crystals with high surface density effectively produces a thick sample. Impurities in the solution often

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produce the same results. In certain cases electrodeposition may be employed to advantage but in general this method requires careful control of conditions which may be different for each substance. It is difficult to obtain satisfactory plates, particularly for electropositive elements such as the actinide elements in which reduction to the metal is not possible.

The most generally acceptable method employed in this laboratory for a wide variety of substances consists of vacuum sublimation. In the presently considered instances a solution of americium or curium chloride is evaporated to dryness on a tungsten filament. Upon raising the temperature to white heat for a few seconds by passing current through the filament, the sample is vaporized onto a 2 mil platinum plate masked by another plate having a rectangular slit 1 x 1/8 inch which defines the sample shape. The mask and collecting plate are placed about 1/8 inch above the filament. The whole system is maintained at a few microns pressure to prevent formation of tungsten oxide which if formed appears as a dark film on the collecting plate. Under optimum conditions the vaporized sample can contain up to  $10 \, \mu \text{g/cm}^2$  of active atoms and still give good resolution, but normally for high resolution the limit is about  $2 \, \mu \text{g/cm}^2$ .

#### Sample Exposure and Track Detection

Referring to Figure 1, the alpha particle beam is defined by the slit and baffle system as indicated. With a uniform magnetic field the width (S) of the image on the photographic plate of a homogeneous beam of alpha particles is

$$2(\Delta S) + r_0^2 a^2 + \cdots$$

where  $\Delta S$  is the defining slit width,  $r_0$  the radius of the normal trajectory

1

and  $\underline{\alpha}$  the half angle of emergence of the beam from the magnetic field. This formula applies to a  $60^{\circ}$  sector magnet with plane surfaces for the pole pieces and the factor "2" arises because the photographic plate is placed at an angle of  $30^{\circ}$  with the trajectory of the alpha particles rather than normal to it.

By proper shaping of the source side of the magnet, the second term of equation (1) can be made negligible with respect to the slit width and this feature is indicated on the diagram by the rounded surface of the magnet. Since the magnetic field is not uniform throughout but falls off near the edges of the magnet, baffles are used as a means of confining the beam to the center. Using a slit opening of 0.018 inch and a 3 inch open. In the ing between the baffles, the alpha beam half-width on the photographic plate for a 6 Mev alpha particle is 2 mm which corresponds to 8 kev.

Sample strengths were in general selected to permit exposures of to 2 days. However, exposures as short as 6 minutes and as long as 16 days have been employed. The lengthy exposures were required to examine the alpha groups present in low abundance in complex spectra. The limiting factor for long exposures is the background of the instrument. Because of the undulating character of the background it was found desirable to have the peak heights at least three times the average background for positive identification. A single field of view in the microscope encompasses 1/4 x 1/4 mm of the photographic plate and a scan of one field width across entire height of the plate (2 inches) gave as background about two alpha tracks for each day exposure. On the low energy side of an alpha peak the apparent background could be considerably higher than this presumably because of the low energy tailing due to absorption in the sample.

As already mentioned, the alpha particles are determined photographically. The detecting plates used were 9 x 2 inch Eastman NTA plates with 25 micron thick emulsions and these were examined under a 450 power microscope with bright field illumination. The track length of a 6 Mev alpha particle is approximately 25 microns and because of the position of the plate as shown in Figure 1, the track makes an angle of 30° with the plane of the emulsion. Because of the small angle of acceptance parallel to the magnetic field permitted by the gap between the pole pieces, the tracks should be nearly parallel and only these are recorded. A photograph of one field of view is shown in Figure 2 in which are seen six acceptable alpha tracks and two tracks which are rejected.

The particular microscope stage employed in these measurements cannot hold a 9 inch plate so each plate was cut into three parts. Before sectioning, an axial line was ruled with a razor blade along the center of the plate parallel to the long dimension. Cross lines perpendicular to the axial line were also ruled in each of the proposed sections. The distances between the intersections of the lines were measured and after sectioning these served as indices to relate distances on the three sections.

In counting the tracks, the microscope stage was moved perpendicular to the axial line giving a scan 1/4 mm wide across the width of the plate. The stage was then moved one field of view parallel to the axial line and another scan made. The count from each scan was plotted on a count versus distance graph as shown in Figure 3.

#### Dispersion

The energy dispersion on the plate for a normal trajectory  $\mathbf{r}_0$  and energy  $\mathbf{E}_0$  is:

Dispersion = 
$$E_0/2r_0$$
. (2)

The relation of the magnetic field to  $E_0$  and  $r_0$  is given by equation (3) in which B is in gauss,  $r_0$  in centimeters and  $E_0$  in electron volts.

$$B = \frac{144}{r_0} E_0^{1/2}$$
 (3)

Since the position of the normal trajectory cannot be determined precisely, it is not possible to determine the value of B necessary to focus a particular alpha particle at that point and  $E_0$  is best eliminated between the equations (2) and (3). The resulting equation (4) for the dispersion is then:

Dispersion = 
$$\frac{r_0 B^2}{2 \times 144^2}$$
 (4)

In addition,  $r_0$  cannot be expected to be precisely equal to the nominal radius of curvature of the magnet (75 cm) because of lack of precision in construction and alignment of source and detector. An effective radius, however, can be determined by measuring distances between alpha groups of known energy. In other words, the dispersion is obtained experimentally for the particular energy range of interest. For this purpose measurements were made of the complex spectrum of Ra<sup>226</sup>, the distance between the main group of Ra<sup>226</sup> and Em<sup>222</sup>, and between Em<sup>222</sup> and Po<sup>218</sup>. The energies taken for Em<sup>222</sup> and Po<sup>218</sup> alpha particles were those given by Briggs<sup>6</sup> (5.486 and 5.998 MeV) and for the two Ra<sup>226</sup> groups, the determination by Rosenblum<sup>7</sup> (4.795 and 4.6105 MeV). Within the limits of experimental error (about 2 percent of the energy differences) the radius so determined was constant and indicated that the nominal 75 cm radius must be increased by 5.4 percent; that is, energy differences calculated by the use of equation (4) using 75 cm radius were low by this amount.

<sup>&</sup>lt;sup>6</sup>G. H. Briggs, Proc. Roy. Soc. <u>A157</u>, 183 (1936).

Rosenblum, Guillot, and Bastin-Scoffier, Compt. Frend 229, 191 (1949).

#### RESULTS

### Am<sup>241</sup> Energy Calibration

Before discussing the complex structure of Am<sup>241</sup>, the energy determination will be mentioned so that the groups may be referred to according to energy. The nuclide Am<sup>241</sup> was that by which the element was first identified<sup>8</sup> and it is prepared in isotopically pure form from Pu<sup>241</sup> decay. The half-life is given as 470 years<sup>10</sup> and 475 years<sup>11</sup> which corresponds to a specific alpha activity of 6.95 x 10<sup>6</sup> disintegrations per minute per microgram. Previous to the present studies the measurement of alpha energy has been made with an ionization chamber coupled to a pulse height discriminator from which the value 5.48 Mev was reported. Although one could observe from the pulse height analysis that the spectrum was not simple, it was not possible to resolve it into its components. The measurement therefore gives the energy of the principal group distorted to an unknown extent by one or more other groups.

The energy for Am<sup>241</sup> was determined with the spectrograph by comparing that of the principal group with two standards, Po<sup>210</sup> and Rn<sup>222</sup>. The other groups of Am<sup>241</sup> were assigned energies by comparison with the main group.

Seaborg, James, and Morgan, <u>The Transuranium Elements: Research Papers</u>,
National Nuclear Energy Series, Plutonium Project Record, Vol. 14B

(McGraw-Hill Book Company, Inc., New York, 1949), p. 1525.

<sup>&</sup>lt;sup>9</sup>Ghiorso, James, Morgan, and Seaborg, Phys. Rev. <u>78</u>, 472 (1950).

<sup>&</sup>lt;sup>10</sup>B. G. Harvey, Abstract of Papers, XII International Congress of Pure and Applied Chemistry (September, 1951), p. 358.

<sup>&</sup>lt;sup>11</sup>Cunningham, Thompson, and Lohr, unpublished data (1950).

 $<sup>^{12}</sup>$ A. Ghiorso, unpublished data (1948).

The comparison between  ${\rm Am}^{241}$  and  ${\rm Rn}^{222}$  is shown in Figure 3. The plate was made by placing sources of  ${\rm Am}^{241}$  and radium simultaneously in the spectrograph for an exposure of 27 hours. That the higher energy group is that of  ${\rm Rn}^{222}$  could be proved by comparing the observed number of tracks with that expected from the observed  ${\rm RaA}^{218}$  tracks caught on another part of the plate. If it was assumed that half of  ${\rm RaA}^{218}$  formed from  ${\rm Rn}^{222}$  left the source by the recoil mechanism, the observed number of tracks in the high energy component was in agreement with the expected amount from  ${\rm Rn}^{222}$ . The resolution of the curve in Figure 3 was aided by a separate exposure with the radium source alone in which the width of the  ${\rm Rn}^{222}$  peak at half maximum was determined. The distance between the peaks was 2.7  $\pm$  0.3 mm which corresponds to an energy difference of  ${\rm ll}^{\pm}$  2 kev. Taking the energy of  ${\rm Rn}^{222}$  as 5.486 MeV, the energy of the principal group of  ${\rm Am}^{241}$  becomes 5.475  $\pm$  0.002 MeV.

Similar exposures were made with Po<sup>210</sup> the results of which are shown in Figure 4. The distance between peaks was 45.8 mm which corresponds to an energy difference of  $179 \pm 2$  kev. From this measurement the main group of Am<sup>241</sup> has an energy of  $5.477 \pm 0.002$  Mev. The energy which we shall use is  $5.476 \pm 0.002$  Mev. As will be described, the most energetic alpha group is higher in energy than this group by 70 kev therefore its energy is 5.546 Mev. The decay energy\* of Am<sup>241</sup> is accordingly 5.640 Mev.

<sup>\*</sup>The term "decay energy" or "alpha energy" refers to the disintegration energy including the recoil energy. The kinetic energy of the alpha particle is explicitly designated "alpha particle energy" or "particle energy."

## Complex Structure of Am<sup>241</sup>

There are now known to be six measurable alpha groups of  $Am^{241}$ , three of which are of greater energy than the most prominent group. In discussing complex spectra, a system for designating the different groups is of value. That used here is directed toward visualizing the energy relations of the groups and was suggested by A. Chiorso. In this system the group known or thought to represent the ground state transition is termed a(0) or, in this case,  $Am^{241}(0)$ . The appropriate energy in kev above the ground state is placed in parentheses for each of the other alpha groups. Thus the most prominent alpha group of  $Am^{241}$  leads to a state 71 kev above the ground state and is designated  $Am^{241}(71)$  or the a(71) group.

The complete alpha spectrum is shown in Figure 5. For these particular data, the source consisted of 2.9 micrograms of Am<sup>241</sup> (2.0 x 10<sup>7</sup> disintegrations/minute) and the exposure was for 94 hours. Because of the disparity in abundance of the different groups, complete peaks cannot be shown to the same scale. In exposing the plate long enough to register a statistically significant number of tracks of the rare groups, too many tracks for counting were registered at the positions of the principal groups. Partial scans across the plate were made for these peaks and the results are also shown in Figure 5. The peak widths at half maximum are essentially the same for all peaks.

The relative abundances of the groups were obtained by counting the tracks of the two most prominent groups on a plate exposed for a shorter period of time and comparing with the rarer peaks from the long exposure. The abundances were virtually the same whether integrated numbers of tracks under the peaks or the peak heights were compared: A summary of the

abundances and the corresponding partial half-lives which will be referred to later are given in Table I. The values in each case represent at least two independent measurements. The sum in abundance of the two highest energy... groups is known with better precision than each group separately because of the uncertainty in resolution. For this reason they are listed both ways in Table I.

Table I. Abundances of Am<sup>241</sup> alpha groups.

					••
Alpha	group <sup>a</sup>	Percentage abundance	<b>2</b>	Partial half-life	- <b>&gt;</b> ****。
		0.57 (=0.0			ing the second s
. *	a(0)	0.23 (±0.0	06 <b>)</b> % %	$2.1 \times 10^{5}$	rational expension and the second
	a(11)	% 0.34, (±0,.0	06)	$1.4 \times 10^{5}$	en en sy kling general in.
	a(43)	0.21 (±0.0	)2)	$2.3 \times 10^5$	ing was transfer and the
	a(71)	84.2 (*1.5	<b>5)</b>	564	
		13.6 ( 1.4			and the second section of the second
	a(170)	1.42 (±0.1	.5)	3.3 $\times 10^4$	A Commence of the Commence of

<sup>&</sup>lt;sup>a</sup>The highest energy group, designated a(0), is taken to represent the ground state transition. The parenthesis-enclosed figures used for the other groups indicate the energy levels in kilovolts above the ground state with which the alpha groups are associated.

The second of th

DBased on 475 year measured half-life.

11

The energy range of the observed peaks was 5.38 to 5.55 Mev. Counting the plate outside of this range revealed that there can be no peak between 5.21 and 5.38 Mev of greater abundance than 0.17 percent and from 5.55 to 5.64 Mev none in more than 0.07 percent abundance. These limits were based on the respective background counts in the regions.

Determinations of energies of the several groups were made relative to the principal alpha group, a(71), which was standardized against  $\mathrm{Rn}^{222}$  and  $\mathrm{Po}^{210}$  as already described. The actual comparisons were made by extrapolating the high energy side of each peak to the intercept after subtraction of the estimated background count and tailing from other groups. The method for obtaining the dispersion in order to translate positions on the plate to energy differences has already been described.

The results of several measurements of alpha particle energies are summarized in Table II. The measured alpha energy differences with their estimated limits of error are as indicated. Also shown are the selected best values and the corresponding energies of the alpha groups based on 5.476 Mev for the most prominent group. In the last column are shown the energy levels above the ground state of Np<sup>237</sup> with which each alpha group is associated. These levels are obtained by correcting the differences of alpha group energies for the corresponding differences in alpha decay recoil energy. It is differences between these numbers which should correspond to gamma ray energies.

<sup>\*</sup>Series  $\underline{\underline{a}}$  and  $\underline{\underline{b}}$  refer to independent counts of the alpha tracks on the same plate.

## Decay Scheme of Am<sup>241</sup>

The complexity of the decay scheme of Am<sup>241</sup> may be visualized readily from the number of observed levels. The precise measurement of some of the gamma rays and L-series x-rays with a bent crystal spectrometer is covered in another paper.<sup>13</sup> The L x-rays arise from internal conversion processes of the gamma rays and they are identified by matching observed energies with those predicted for neptunium according to extrapolations of the Moseley relationship. It is presumed that other observed photons are nuclear gamma rays and an attempt is made to match these with energy levels deduced from the alpha particle spectrum.

Figure 6 shows a partial decay scheme with the energy levels shown corresponding to the alpha particle spectrum. Only a part of the measured gamma rays have been entered because the positions of all relative to the energy levels defined by the alpha spectrum are not uniquely determined. Matching of intensities of gamma transitions would be a valuable guide but at this time the conversion electron spectrum and abundances have not been measured. The discussion of all of the gamma rays and their possible placement in a decay scheme will be found elsewhere but a few features of the gamma ray spectrum are worth mentioning here.

The most abundant gamma ray has an energy of 59.78 kev and corresponds closely with the transition from the state reached by the most abundant alpha group, a(71), to the 10.8 kev level. This gamma ray had been reported initially by Seaborg, James, and Morgan<sup>8</sup> with an energy of 62 kev based on absorption measurements. The conversion electron spectrum of Am<sup>241</sup> has

<sup>13</sup>C. I. Browne and I. Perlman, Phys. Rev. (to be published).

been measured by 0'Kelley who found  $L_{\rm I}$ ,  $L_{\rm III}$ ,  $L_{\rm III}$ , M and N lines corresponding to a gamma ray of 59.4 keV. Conversion lines of softer gamma rays could not be resolved from each other and from the Auger electrons.

The gamma ray of 59.78 kev is matched precisely by the sum of two other gamma rays of 26.43 and 33.36 kev and these in turn agree with differences between levels excited by a(71) and a(43), and by a(43) and a(11), respectively. It is interesting to note that no gamma ray from a known energy level to the ground state has been observed. The de-excitation of the 10.8 kev level would not have been measured even though a significant fraction were to go by a radiative transition. Two other gamma rays have been entered in Figure 6 with broken lines to indicate the transitions. These same lines in conjunction with others could be given other equally good assignments and are entered as shown only to illustrate that in any case an energy level must be postulated for which there is no measured alpha group. In view of the fact that several of the alpha groups can barely be detected it would not be surprising if there were one or more which are in too low abundance.

Further fragmentary information on the decay scheme of Am<sup>241</sup> has been obtained in this laboratory and will be discussed in the paper dealing with the gamma ray spectrum.

## Cm<sup>242</sup> Energy Calibration

The highest energy group of  ${\rm Cm}^{242}$  is also the most abundant and has been used in the energy determination. This nuclide with 162.5 days half-life<sup>15</sup> was first prepared by alpha particle bombardment of  ${\rm Pu}^{239}$  in

<sup>14</sup>G. D. O'Kelley, Ph.D. Thesis, University of California, 1951.

<sup>&</sup>lt;sup>15</sup>Hanna, Harvey, and Moss, Phys. Rev. 78, 617 (1950).

the cyclotron<sup>16</sup> but is more readily obtained in quantity by neutron irradiation of  $Am^{241}$ . The best energy determination using the ionization chamber and pulse analyzer method is 6.08 Mev.<sup>12</sup>

In the present measurements with the alpha particle spectrograph,  $\rm Cm^{242}$  was compared with RaA<sup>218</sup> (5.998 Mev) and with the main group of  $\rm Am^{241}$  (5.476 Mev). The method used was similar to that employed for americium (see explanation and Figures 3 and 4). Based on the RaA<sup>218</sup> calibration, the energy is  $6.110^{+}0.003$  Mev and the  $\rm Am^{241}$  comparison gave  $6.112^{+}0.010$  Mev. With the limits of error as given, the close agreement is fortuitous and the value selected is that of the more accurate measurement,  $6.110^{+}0.03$  Mev. Since this group is probably that of ground state transition, the decay energy of  $\rm Cm^{242}$  is accordingly 6.211 Mev.

# Complex Structure of Cm<sup>242</sup>

The spectrum of Cm<sup>242</sup> in the energy range 6.0 - 6.1 MeV is shown in Figure 7. Aside from the two groups shown here none has been found between 5.5 and 6.5 MeV but the limits of detection vary with position. These limits are best shown graphically as in Figure 8 which illustrates the low limits of detection (<0.01 percent) on the high energy side of the main peak.

Several exposures have been made to determine the energy differences between the two alpha groups and their relative abundances. The data are summarized in Table III. The average difference in energy is 45.7 kev which, taken with the energy of 6.110 Mev for the main group, makes the low energy group 6.064 Mev. The abundances of the two groups are 73

<sup>16</sup> Seaborg, James, and Ghiorso, <u>The Transuranium Elements: Research Papers</u>, National Nuclear Energy Series, Plutonium Project Record, Vol. 14B (McGraw-Hill Book Company, Inc., New York, 1949), p. 1554.

and 27 percent, and from the measured half-life of 162.5 days, the partial half-lives for the two groups are 222 and 602 days, respectively.

The gamma ray corresponding to the 46.5 kev transition has been detected in low yield\* and low energy electrons have been found in abundance.

O'Kelley has measured a series of conversion lines corresponding to a gamma ray of about 43 kev.

Table III.	Energy	differences	and	abundances	of	the
*	alpha	groups of Cm	242.			•

Experiment	Differences of	Separation	Abundances		
number	alpha particle energies	between energy levels	Low energy group (%)	High energy group (%)	
4-8	45.9 ± 0.7	46.6 ± 0.7	27 ± 2ª	73 <sup>+</sup> 2 <sup>a</sup>	
4-16	45.2 <sup>±</sup> 1.0	46.0 ± 1.0	26.6	73.4	
4-20	46.0 ± 0.9	46.8 ± 0.9	26	74.	
4-80	45.8 <sup>±</sup> 1.3	46.6 ± 1.3	27	73	
avera	ges 45.7	46.5	27	73	

<sup>&</sup>lt;sup>a</sup>Abundances determined by integrating alpha track counts under peaks; others determined by comparing peak heights.

<sup>\*</sup>In this laboratory, A. Ghiorso has observed this gamma ray with a proportional counter coupled to a pulse height analyzer and D. F.

Martin has measured it with a scintillation counter spectrometer.

#### ALPHA DECAY THEORY

The theory of the alpha decay process relates the four factors: decay constant, nuclear charge, decay energy and nuclear radius. A fifth parameter, the internal potential of the alpha particle, is usually eliminated in the solution of the equations. Of these, only the radius cannot be determined with accuracy by measurement but can be calculated insofar as the theory is valid if the other three parameters are known. Any shortcoming of the theory in describing a particular alpha decay process is, of course, reflected in the calculated radius. It has become a basic precept in nuclear theory that nuclear volumes are not expected to undergo wide variations from the simple addition of the number of nucleons contained. If the shapes of the nuclei, or more precisely the charge distributions, do not differ much in the limited region under consideration, then the radii too should vary in a regular manner. From these considerations it is seen that regularity of calculated nuclear radii may serve as a check on the alpha decay theory. It has been found that the even-even alpha emitters do indeed give remarkably uniform nuclear radii calculated from one-body theory. 3,4,17 The form which the expression for nuclear radius takes is simply  $r = r_0 A^{1/3} \cdot 10^{-13}$  cm. The radius parameter  $r_0$  varies somewhat with The theory followed here was developed the particular theory employed. by Preston, 18 elaborated by Kaplan, 7 and also includes a correction term for the alpha energy pointed out by G. Ambrosino and H. Piatier. 19 This correction amounts to an addition of 40 kev in the case of curium and is

<sup>17&</sup>lt;sub>I</sub>. Kaplan, Phys. Rev. <u>81</u>, 962 (1951).

<sup>&</sup>lt;sup>18</sup>M. A. Preston, Phys. Rev. <u>71</u>, 865 (1947).

 $<sup>^{19}</sup>$ G. Ambrosino and H. Piatier, Compt. rend. 232, 400 (1951).

the difference in binding energy of orbital electrons in curium and plutonium.\*

There are a number of reasons for wanting a convenient method for checking alpha decay data with the theory and since the calculations are tedious, a graphical representation has been evolved.  $^{20,21,3}$  In this a family of curves are plotted which are derived from the theory applied to the even-even alpha emitters. First, values of  $\underline{r_0}$  are obtained from values of decay energy, half-life and charge for those even-even nuclides for which reliable data are available. The best value of  $\underline{r_0}$  is taken and this defines the normal nuclear radius for each mass number. Using this value one calculates the half-life corresponding to the measured alpha energy for each nuclide. For each element a smooth curve is obtained on a half-life vs. energy plot as shown for plutonium and curium in Figure 9. The curve for americium is interpolated. The measured half-life for an alpha emitter is then entered at the appropriate energy. From the manner in which the curves were constructed it is seen that they should be the best curves drawn through the even-even nuclides.

The utility of such curves lies in the ease with which any new alpha emitter can be tested for agreement with the theory simply by observing the departure from its curve. If a point lies above its curve, the half-life is longer than expected and the decay process is said to be hindered. Now that it is found that virtually all alpha emitters have complex structure, these curves supplant a large number of calculations required to determine the degree of hindrance for the individual alpha groups. Such

<sup>\*</sup>A further small correction for the screening effect of orbital electrons on the potential barrier has not been included and probably should be considered in further refinements.

 $<sup>^{20}</sup>$ A. Berthelot, J. phys. radium VIII  $\underline{3}$ , 52 (1942).

<sup>&</sup>lt;sup>21</sup>S. Biswas, Indian J. Phys. <u>23</u>, 51 (1949).

comparisons for  $Am^{241}$  and  $Cm^{242}$  are shown in Figure 9 and will be discussed further presently.

Another great utility of these curves is in predicting half-lives of new species being sought. It is possible to predict an alpha energy with considerable accuracy<sup>3</sup> and from this value it is simple to read from the curve the minimum alpha decay half-life. In the case of <u>even-even</u> nuclides experience has shown that these half-lives are quite accurate and for other nuclear types the actual half-lives are usually several fold greater than those read from the curves.

Returning to the curves of Figure 9, a brief statement should be given for the choice of  $\underline{r_0}$  = 1.51 which was used in their construction. This has turned out to be the best value for the <u>even-even</u> nuclides of plutonium and curium for which data are available. The inclusion of a wider range of elements and the refinement of measurements may well necessitate revision of this value. Furthermore, although a great simplification results from the assumption of a single value of  $\underline{r_0}$  to fit all alpha emitters in this region, this obviously cannot be rigorously correct and as data are refined it may become advantageous to let  $\underline{r_0}$  vary slowly. If  $\underline{r_0}$  varied erratically from nuclide to nuclide, the construction of a simple family of curves such as dealt with here would not be possible and indeed such behavior is encountered in the region of 126 neutrons. 3,4

It is seen that the ground state transition of  $Cm^{242}$ , a(0), lies on the curium curve which fact indicates its accord with other curium and plutonium nuclides. The lower energy group of  $Cm^{242}$ , a(47), lies above the curve in degree corresponding to a half-life ~1.5 times longer than expected from the theory. What selection rules are responsible for this

slightly hindered decay are not yet formulated.

The disposition of the  ${\rm Am}^{241}$  alpha groups is decidedly different. Here, the three highest energy groups, designated a(0), a(11), and a(43), are highly hindered and have half-lives roughly 1000 times longer than the simple theory would demand. On the other hand, a(71) follows closely the pattern of an <u>even-even</u> nuclide and as a result is the most abundant alpha group of  ${\rm Am}^{241}$  even though it is only the fourth highest in energy. The two alpha groups of still lower energy are again hindered although in lesser degree than the three highest energy groups.

There is at present no quantitative explanation to account for the degrees of hindrance of the various alpha groups. It has been pointed out<sup>3</sup> and Preston<sup>17,22</sup> has demonstrated that no explanation to include such high degree of hindrance as for several of the Am<sup>241</sup> groups is likely to come from spin changes in the alpha transitions. An hypothesis which we shall consider further is that the delay is involved in assembling the components of the alpha particle and that the quantum states of the affected nucleons are involved.

#### ACKNOWLEDGMENTS

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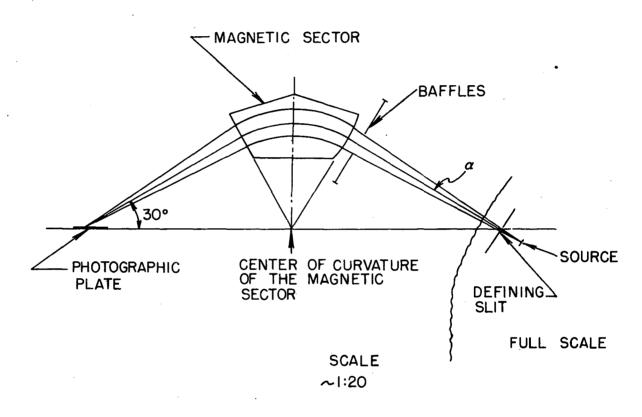
<sup>&</sup>lt;sup>22</sup>M. A. Preston, Phys. Rev. <u>83</u>, 475 (1951).

#### Figure Captions

- Fig. 1 Schematic diagram of the optics of the spectrograph.
- Fig. 2 Alpha tracks in photographic emulsion.
- Fig. 3 Positions of  $Em^{222}$  and the principal  $Am^{241}$  alpha group. Dispersion - 3.96 keV/mm
  - spectrum as measured
  - ——— Em<sup>222</sup> peak resolved by subtracting background and using half-width for the peak as determined on a separate measurement with the Em<sup>222</sup> sample alone
  - resolved alpha group of Am<sup>241</sup>
- Fig. 4 Positions of Po<sup>210</sup> and the principal Am<sup>241</sup> alpha group.

  Dispersion 3.90 kev/mm
- Fig. 5 Alpha spectrum of Am<sup>2/41</sup>.
- Fig. 6 Partial decay scheme for Am<sup>241</sup>. Energy levels shown were obtained from alpha spectra. Gamma ray energies shown were obtained with a bent crystal spectrometer.<sup>13</sup>
- Fig. 7 Alpha spectrum of  $C^{242}$ .

  Dispersion 4.01 keV/mm.
- Fig. 8 Upper limit of abundance of alpha groups of  $Cm^{24/2}$  as a function of energy (arrows indicate positions of known alpha groups).
- Fig. 9 Half-life energy relations of  ${\rm Cm}^{242}$  and  ${\rm Am}^{241}$  alpha groups.



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Fig. 1

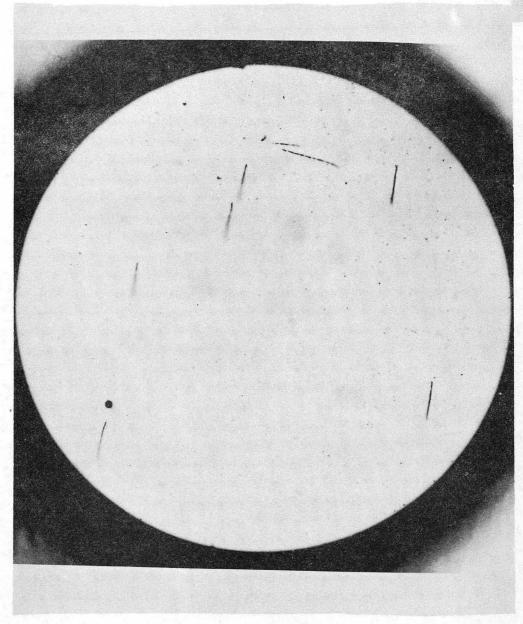


Fig. 2

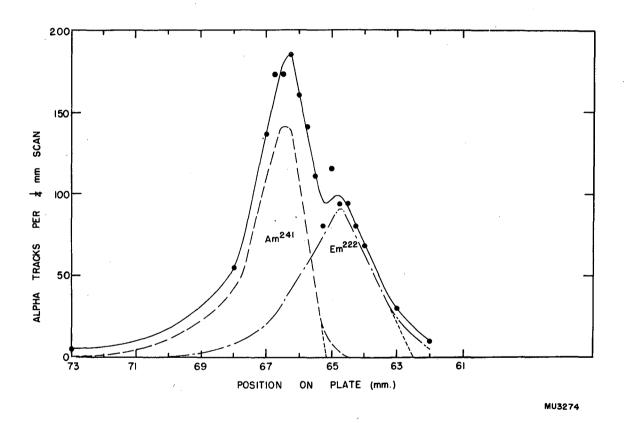


Fig. 3

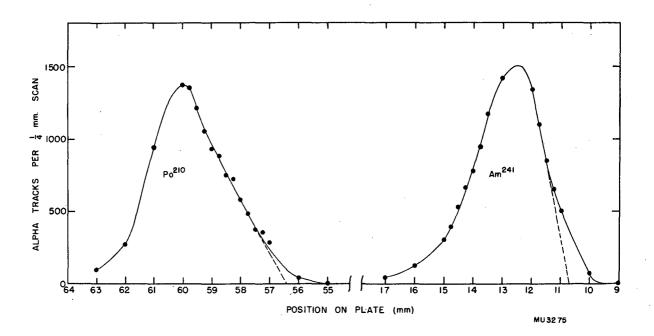


Fig. 4

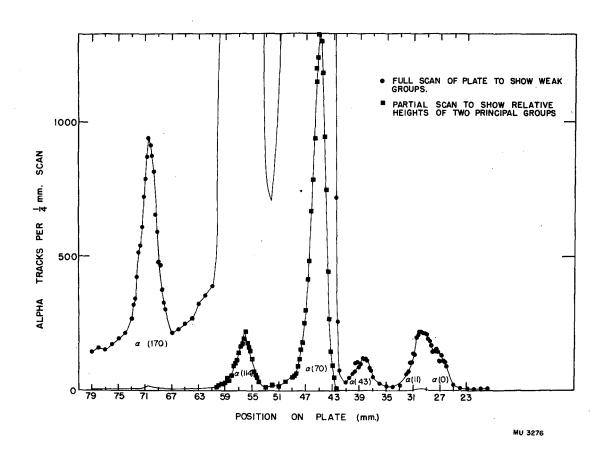


Fig. 5

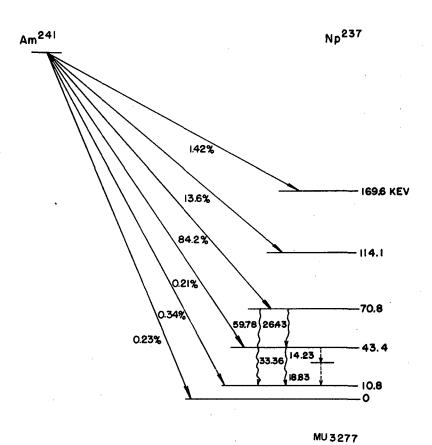


Fig. 6

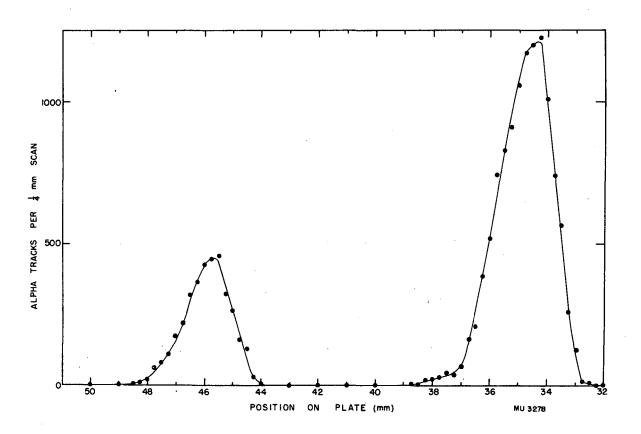


Fig. 7

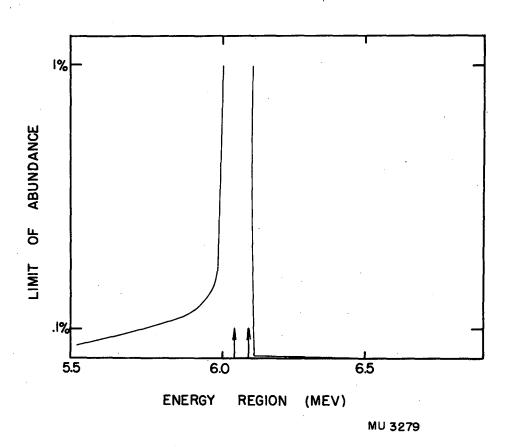


Fig. 8

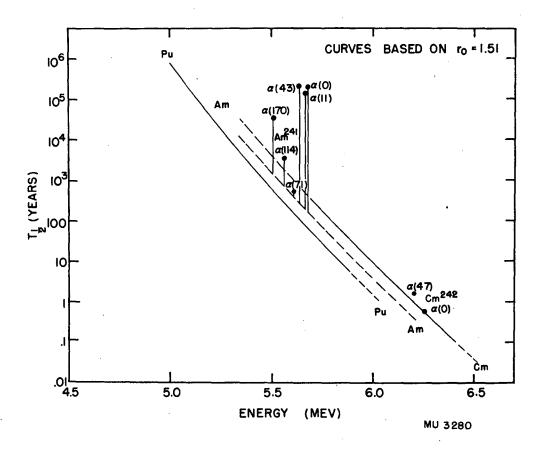


Fig. 9