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ALPHA RADIOACTIVITY

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May 24, 1954

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INTRODUCTION

The comparison of the number of alpha emitters known at present with those known a few years ago serves as a rough index of the comparative research activity on alpha radioactivity and associated problems. By 1944 just 26 alpha emitters had been definitely identified and reported, and of these all but one were members of the natural radioactive families. (Some others were found during the war years but publication was delayed.) In 1948 the total number of alpha emitters had climbed to nearly 90 and at the end of 1953 there were more than 160 known. The increase in numbers of alpha emitters has been accompanied by diversification of the information attainable by the treatment of alpha decay data as well as a better insight into those features of alpha-radioactivity already recognized. It is perhaps worth recalling, however, that the basic alpha decay theory (1, 2) was developed in the "days of the natural radioactivities" and precise absolute alpha energies were determined during this period (3).

The huge increase in numbers of alpha emitters is a result of several stimuli. The wartime military efforts in atomic energy were directly concerned with several new alpha emitters and as a result of their preparation a number of others appeared as by-products. This work stimulated the desire to know the nuclear properties of other isotopes of these elements and developed into a search for higher transuranium

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[1] The survey of the literature pertaining to this review was concluded in April, 1954.

elements as well. It is interesting to note that at present there are as many elements known above uranium as there are between uranium and bismuth. Another important source of alpha emitters came with the construction of particle accelerators operating in a new high energy range. With the use of such instruments (particularly the Berkeley 184-inch cyclotron), exploration into the region of neutron deficient isotopes became possible and resulted in the discovery of a wide range of new alpha emitters including a group among the rare earth elements.

This review will be confined in large measure to the development of a few topics which have stemmed from the investigations of the past several years. The classical work on alpha radioactivity will not be covered and no special treatment will be given to the natural alpha emitters.

A few brief statements outlining these newer developments are given in the following:

1. <u>Regularities in alpha-decay energy.</u> -- It is possible to correlate decay energies on a semi-empirical basis and the observed regularities make it possible to predict rather closely the decay energies of unknown species.

2. <u>Regularities of decay-lifetimes.</u> -- It is now known that one-body alpha decay theory, not far different in form from that originally proposed, is capable of yielding precise lifetime values for most even-even nuclei but not for other types. Lifetimes can be predicted on a semi-empirical basis for almost all cases.

3. <u>New developments in alpha decay theory.</u> -- The results to be cited on odd nucleon types and on the complex spectra of all types serve to delimit the applicability of present alpha decay theory and to define the problems which must be solved by a more comprehensive theory. Some

of the attempts to build upon the classical theory will be mentioned.

4. Spectroscopic states deduced from alpha decay. -- Almost all alpha emitters show complex spectra when examined with instruments of high resolution. The mapping of nuclear spectroscopic states populated in the alpha-decay process has yielded substantial information on the general picture of energy levels in the heavy element region.

Unfortunately, it will not be possible in this review to discuss the important advances in detection and energy determination of alpha particles. Also omitted are the means for producing the many alpha emitters and any individual discussion of many of the interesting new species.

ALPHA DECAY ENERGY

Somewhat beyond the middle of the periodic system alpha emission becomes energetically possible for all nuclides along the band of beta stability. Alpha decay lifetimes are, however, strongly dependent upon alpha decay energies, and only when the decay energy exceeds a certain value do the lifetimes come down into the range where radioactivity can be detected. Since the potential barrier against alpha decay increases with atomic number, the "critical" alpha decay energy increases with atomic number. As a rough gauge of this effect we may say that the energy required for a half-life of 10^{12} years is 2 Mev in the region of samarium (atomic number 62) and 4 Mev in the region of uranium (atomic number 92). The energy dependence in the heavy element region is such that doubling the energy (4 Mev to 8 Mev) decreases the half-life by a factor of more than 10^{20}.

The general energy trends along the line of maximum beta stability are indicated in Fig. 1 which is similar to a drawing published

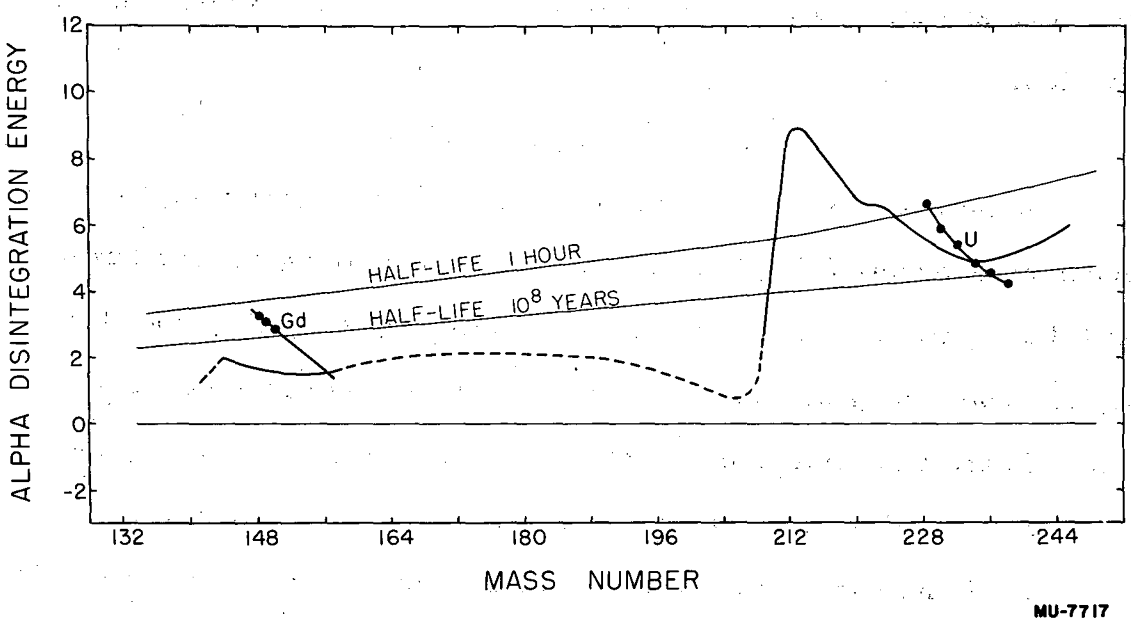

Fig. 1. Alpha decay energy profile.

Broken line portion of curve indicates region where direct alpha decay measurements are absent. Plotted points on segments of crossing curves pertain to known alpha emitters of gadolinium and uranium. Half-life guide lines indicate positions of applicable lifetimes as a function of mass number.

by Kohman (4). The curve was constructed by making use of available mass data in the region in which alpha emission is not discernible. Aside from the smooth curve showing alpha decay energies, three other lines are shown. One of these simply shows the line of demarcation between stability and instability toward alpha emission and the other two show the approximate positions at which half-lives would be 10^8 years and 10^{-4} years (about 1 hour). As mentioned, the smooth curve goes through the center of beta stability for each mass number. If we also consider neutron deficient isotopes of each element, it will be found that their alpha decay energies are greater, a point which will be considered in more detail presently. Two such series of isotopes (for gadolinium and uranium) are indicated by points and segments of curves crossing the main curve. The position of shell closures at 82 protons and 126 neutrons is readily seen through its influence on alpha decay energy.

The mass data available are not nearly accurate enough to define the curve of Fig. 1 with the precision required to be useful as a guide for alpha decay properties. Uncertainties of several Mev are present in most of the data while a fraction of a Mev is sufficient to change a lifetime from the region of easy observation to that of extremely long periods. The curve is consistent with known alpha decay properties and can be altered in detail as more precise data become available.

Summary of alpha energies.--A compilation of all alpha energies in the heavy element region is given in Fig. 2.[2] The objective of this

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[2] Most of the references for data shown here and in Fig. 3 will be found in the 1953 Table of Isotopes (reference 25). For newer data consult the following: Po^{206} (5), Em^{211} (6), Em^{218} (7), Em^{220} (8),

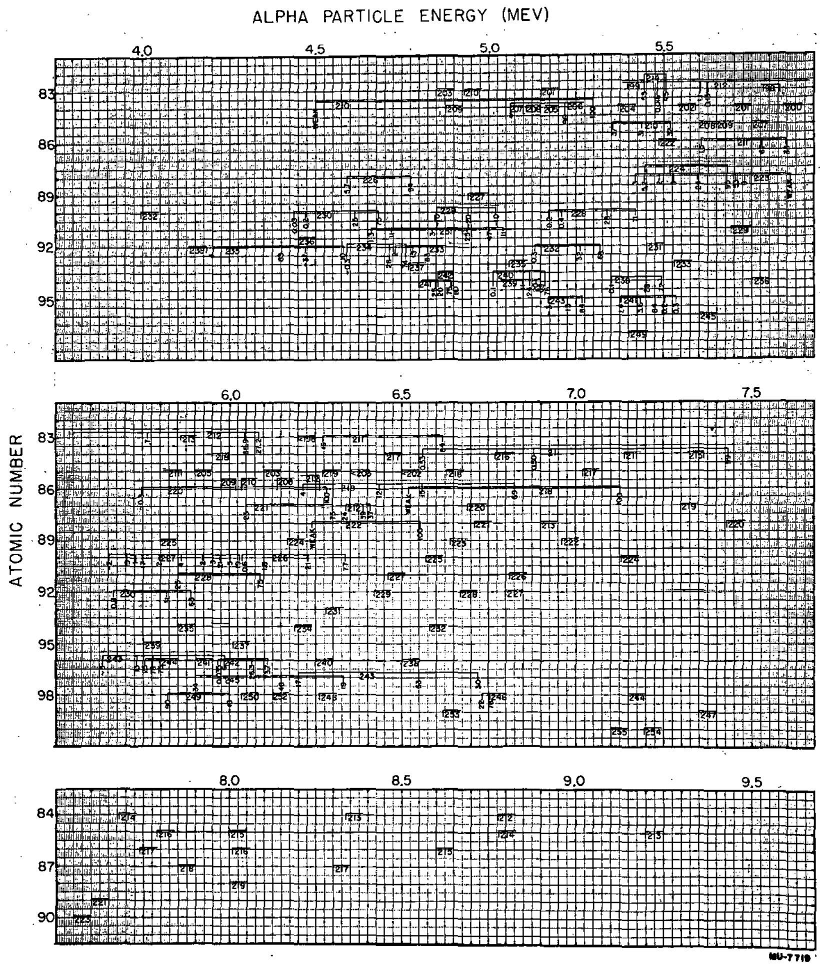

Fig. 2. Alpha particle energies and abundances.

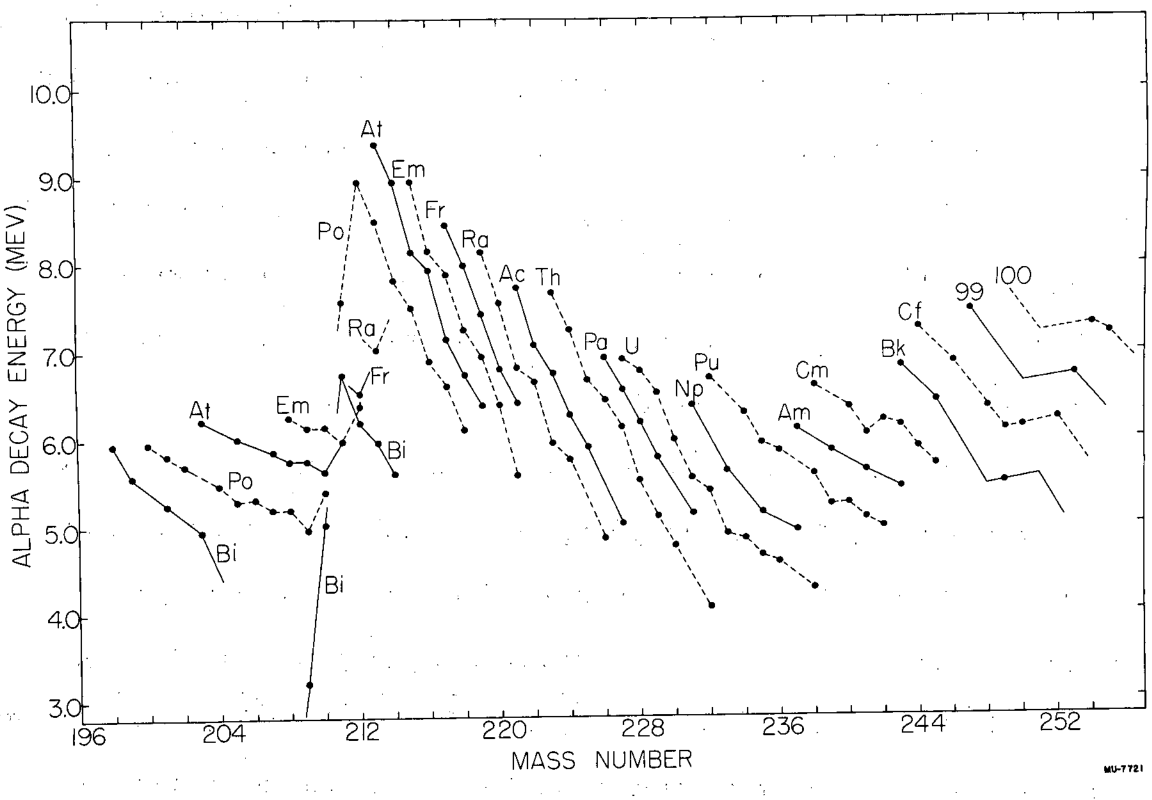

Fig. 3. Alpha decay energy vs. mass number.

Ra^{222} (7), Ra^{224} (9), Th^{226} (7), Th^{228} (9), Th^{230} (10, 113), Th^{232} (11), U^{230} (7), U^{232} (8), U^{234} (12), U^{238} (11), Pu^{238} (13), Pu^{240} (8), Pu^{241} (8), Pu^{242} (8), Am^{241} (14), Am^{243} (14), Cm^{243} (15, 8), Cm^{241} (16), Bk^{249} (17), Cf^{246} (18), Cf^{248} (19), Cf^{249} (20, 19, 17), Cf^{250} (20, 19, 17), Cf^{252} (20, 19, 17), 99^{247} (21), 99^{253} (20, 19, 22, 23), 100^{254} (24, 23, 22, 19), and 100^{255} (19).

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particular presentation is to show alpha spectra on a sufficiently expanded scale to permit reading to the nearest few kilovolts. The short vertical lines indicate the energy values of alpha groups and those which comprise the spectrum of a particular alpha emitter are joined by a bar with the mass number indicated. The per cent abundance of each group which is part of the complex spectrum is noted at the energy position of the group. No abundance designation is given where only a single group has been observed.

Figure 2 may also be used by those measuring alpha spectra as a ready reference to all alpha groups of a particular energy. For example, if a new alpha spectrum under examination shows a weak alpha group at 5.50 ± 0.05 Mev, one must show that the group does not belong to Bi^{199}, Bi^{212}, Bi^{214}, At^{210}, Em^{222}, Ra^{223}, U^{231}, Pu^{238}, Am^{241}. Often the mode of preparing the sample can be used to judge most of the possibilities. For others, a search can be made for other components of the spectra which would be present and these must also pass the test of having the proper abundances.

The energy values shown in Fig. 2 were obtained by a number of different methods which are inherently different in precision. Since there appeared to be no convenient method for expressing limits of error for each datum in Fig. 2, it will be necessary to consult the original papers for this information. Most energies determined are not absolute but depend upon comparison with standards, so a list of some of the most accurate values (absolute and comparative) suitable for standards are shown in Table 1.

Alpha decay energy systematics. -- The value of a curve such as that in Fig. 1 is to help visualize in broad outline the conditions and regions of alpha instability. A good deal more is to be learned from a more detailed examination of the regions where alpha radioactivity is prominent. It is observed that alpha decay energies[3] fall into well behaved patterns, the recognition of which has proved considerably useful for a number of purposes. Two of the functions of these correlations will be mentioned.

Alpha decay series (containing relatively few beta decay steps) provide a continuous series of energy increments from the region of the element lead to the highest elements possible. The many decay

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[3] Alpha decay energy or simply decay energy will refer to the Q value for the alpha emission process and is therefore immediately transferable into a mass or energy difference between ground states of parent and daughter. The actual alpha particle measured for the ground state transition is of appreciably lower energy because of the energy carried off by the recoil atom. Of course, the alpha groups leading to excited states are of still lower energy. The measured energies of alpha groups will be referred to as alpha particle energies or particle energies to distinguish them from decay energies.

Table 1

Absolute Alpha Particle Energy Determinations Including Some Values Measured Relative to RaC'*

Alpha emitter	Briggs(3)	Sturm and Johnson(27)	Rosenblum, Dupouy(28)	Lewis, Bowden(29)	Collins, McKenzie, and Ramm(30)	Best value, Briggs(26)
RaC' (Po^{214})	7.680	7.683				7.680
ThC' (Po^{212})	8.776†		8.780	8.776†	8.786	8.780
Po^{210}		5.298	5.299	5.299†	5.305	5.301
ThC (Bi^{212})α_0	6.082†		6.087	6.082†	6.090	6.086
ThC (Bi^{212})α_{40}	6.043†		6.048	6.042†	6.050	6.047

*All measurements cited were made with magnetic spectrographs except those of Sturm and Johnson (27) who used an electrostatic device. The values listed for the magnetic measurements were corrected as done by Briggs (26) in terms of slight revisions of fundamental constants used in the conversion of Hρ to energy.

†Measured relative to RaC'.

chains either combine through decay or can be related energetically by measured neutron binding energies. By analyzing these data, it is possible to obtain a rather detailed picture of the energy surface[4] in the heavy element region. The shape of the energy surface is readily delineated from these data and regions of greater or lesser stability stand out clearly. A schematic representation of such a surface, exaggerated for purposes of illustration, was given by Perlman, Ghiorso, and Seaborg (31). A complete and critical compilation of all decay energy data and the atomic masses calculated from these has been made by Seaborg and Glass (32). The data include masses of nuclides not yet prepared and these were derived from decay energies estimated by methods which will be indicated below.

Of great value to the experimentalist has been the ability to predict alpha energies and the agreement between predicted and measured values often serves as a criterion for isotopic assignment. Furthermore, if the energy can be predicted, the half-life can be calculated. The forecasting of both alpha energy and half-life has been of inestimable value in the preparation and identification of higher and higher elements.

A number of systems for correlating alpha decay energies have been employed and perhaps that most widely used is illustrated in Fig. 3. Here the isotopes of each element on a mass number versus energy plot are joined, resulting in a family of curves which over a wide region comprise a series of nearly parallel lines. It will be noted that

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[4]Energy surface refers to a surface defined by the energy contents or masses of the nuclides.

in this region (above mass number about 212) alpha energies decrease with increasing mass number for each element; that is, with increasing neutron number. This system in some modification has been used by a number of workers, (33 to 37, 31) while others have used contour map presentation (38, 39) and still different methods (40).

Before turning to the regions in Fig. 3 where sharp changes in the lines occur, let us consider the broad region of regular behavior. The semi-empirical method of calculating atomic masses of Weizsäcker (41) as developed further by Bethe and Bacher (42) and by Bohr and Wheeler (43) pictures the energy surface as a trough having parabolic sections at constant mass number, A. The bottom of the trough goes through the beta stable nuclides and if the energy coördinate is expressed in terms of mass defect, the line along the bottom becomes the familiar "best" mass defect curve. It can be shown graphically (31) or analytically (4) that the observed trend of alpha energy with mass number (Fig. 3) is to be expected from this picture. Differences in slope and spacing of the lines can be interpreted in terms of departures from extreme regularity of the trough such as small changes in curvature of the trough or of its slope.

The dramatic inversion in the alpha energy trend around mass number 212 is a consequence of the major closed shells in this region (44, 37). We can see what happens more precisely by following the curve for the polonium isotopes. From Po^{218} down to Po^{212} the curve follows the normal trend; then Po^{211} is seen to have considerably lower alpha energy than Po^{212}. This can readily be shown to be a reflection of the fact that the binding energy of the 126th neutron in lead (Pb^{207}-Pb^{208}) is greater than that of the 128th neutron in polonium (Po^{211}-

Po^{212}). Similarly, since the binding energy of the 125th neutron in lead (Pb^{206}-Pb^{207}) is greater than that of the 127th neutron in polonium (Po^{210}-Po^{211}), the alpha energy of Po^{210} is lower than that of Po^{211}. After the neutron shell of 126 is well passed and the neutron binding energies change monotonically for both parents and daughters, the regular trend of increasing alpha energy with decreasing neutron number is resumed (see region between Po^{208} and Po^{203}).

The curve for bismuth is seen to parallel the polonium curve with a wide energy spacing between them. This energy spacing is presumably a consequence of the 82 proton shell. The reappearance of alpha radioactivity in highly neutron deficient isotopes of bismuth (45) was the clue needed to establish the generalness of this effect of crossing the region of 126 neutrons (44-52).

It will be noted that the alpha energies for the isotopes with 128 neutrons would be expected to become progressively greater for each higher proton number. Half-lives will accordingly be very short in this region so that preparation and identification of such nuclides would be difficult. However, more neutron deficient isotopes should be more stable just as Po^{210} is more stable than Po^{212}. Such a region has been found for astatine, emanation, and even for radium, showing that the effect of 126 neutrons extends at least this high (51, 52). Undoubtedly these points shown on Fig. 3 join to those of higher neutron number by going through sharp peaks higher than those shown for polonium and astatine.

The question arises as to whether or not there is evidence for other closed shells or subshells on the basis of alpha decay data. A situation similar to that at 126 neutrons, but considerably more subdued, seems to occur at neutron number 152 (19). The evidence comes principally from the alpha energy trend of californium isotopes (element 98) as seen in Fig. 3 showing that the californium isotope with 154 neutrons has a higher energy than those of the next few isotopes of lower mass number. The fragmentary data on elements 99 and 100 are not out of line with this concept and the lines are drawn in Fig. 3 according to expectations. As will be explained later, the positions of closed shells can be correlated with energy level spacings between certain excited states and their ground states, but in this respect the single nuclide known with 152 neutrons (Cm^{248} from alpha decay of Cf^{252}) does not reflect a closed shell configuration (53, 54).

It will be noted in Fig. 3 that the energy increments from isotope to isotope along many of the curves are not very uniform. Aside from the marked inversion in trend in the region of lead and the probable small inversion in conjunction with 152 neutrons, it is seen that in some places the isotopes seem bunched and in others relatively spread out. It has been suggested that there are subshells at 92 protons (55, 56) and at 88 protons (56) in explanation of some of these irregularities. Other inferences of alpha decay energies on possible subshells have been discussed by Broneiwski (57).

Decay energies from energy-balance cycles. -- As a guide in research on new heavy isotopes, the ability to predict decay properties is of great value. By interpolation and extrapolation, the curves of Fig. 3 may be used to estimate alpha energies and these as well as measured values can be used in constructing a self-consistent system of energy-balance cycles which can be used in turn to calculate degrees of β^- instability and other alpha energies.

To illustrate this method of correlating decay energies (32), a segment of the decay cycle representation of 4n + 1 type[5] nuclei is shown in Fig. 4. A few examples of the uses of these cycles will be mentioned. It is noted that by making use of three measured decay energies the alpha energy of the 6.8 day β^--emitter U^{237} is calculated to be 4.25 Mev. This is almost identical with the decay energy of U^{238} which has a half-life of 4.5×10^9 years and the partial alpha decay half-life of U^{237} would be expected to be at least as long. Accordingly, the alpha branching of U^{237} would be only of the order of 10^{-12} so that this mode of decay would be most difficult to observe. On the contrary, the alpha branching of Pu^{241} was similarly estimated to be about 10^{-5} which was within reach for measurement and the alpha energy as subsequently measured is shown in Fig. 4.

Another use of these cycles has to do with predictions of β-stability. If one considers the possibility that Cm^{245} is a β^--emitter and that its preparation would therefore also produce Bk^{245}, the idea should be rejected because it is seen that Bk^{245} is unstable with respect to Cm^{245} by about 0.7 Mev. The estimated alpha energy of Cm^{245} which goes into this calculation cannot possibly be in error by an amount to reverse this conclusion.

An extension of these cycles to still higher elements gives a means of making predictions into a region where measurements have not been made and these predictions serve as an important guide to the experiments. Other cycles can be devised to join different nuclear

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[5]The type "4n + 1" means that all mass numbers are divisible by 4 with remainder 1. All nuclei connected by α- and β-decay processes do not change type.

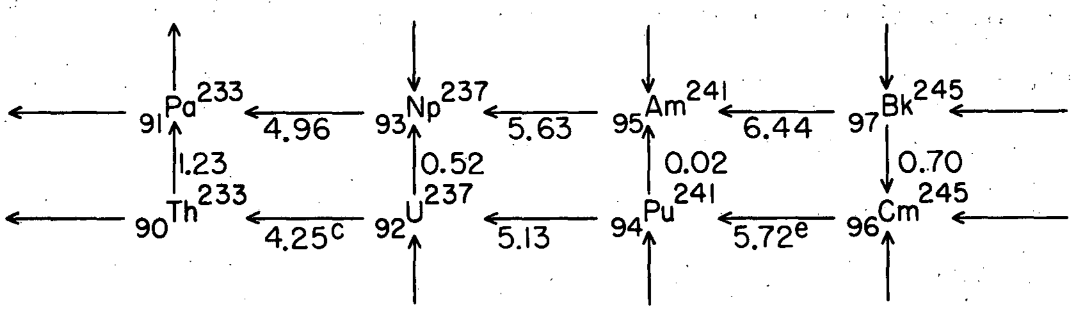

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Fig. 4. Decay cycles for part of 4n + 1 family.

Code: ← alpha decay, ↑ negative beta decay, ↓ electron capture decay. Numerals indicate energies in Mev; those with superscript "c" are calculated by closing the cycles; those with superscript "e" are estimated as by the use of Fig. 3; those unmarked are measured values.

types through measured binding energies. With a single neutron binding energy measurement joining two series, other neutron binding energies can be calculated (32).

Alpha emitters just below lead. -- The removal of neutrons from any element increases the potential toward alpha decay and this is the basis for the main trend in Fig. 3. Alpha active nuclides of gold and mercury have been prepared by removing many neutrons from the stable isotopes (58). In the case of gold, the stable isotope Au^{197} is estimated to have an alpha decay energy of only 1 - 2 Mev, while the isotope observed with an alpha energy of 5.1 Mev is believed to lie in the mass number range 183-187. As neutrons are removed, successive isotopes become more unstable toward orbital electron capture also, but since alpha decay lifetimes are extremely sensitive to energy, this mode of decay should at some point become discernible.

One other alpha emitter in this region has been reported (59), a component of natural tungsten. The extremely low specific activity seems out of line with the measured energy (3.2 Mev) if the emitter has the abundance of one of the known tungsten isotopes, so the authors postulate the existence of a rare isotope (W^{178}) in the natural mixture in amounts too small to detect by mass spectroscopy.

Rare earth alpha emitters. -- Among the rare earth elements we pass through a region where stable or slightly deficient nuclides can decay by alpha emission to the closed shell of 82 neutrons. Such a nuclide with 84 neutrons is Sm^{146} which is beta stable but missing in nature because of its relatively short alpha half-life ($\sim 5 \times 10^7$ years) (60). The alpha energies are summarized in Fig. 5 and although the curves are fragmentary as compared with those in the heavy element

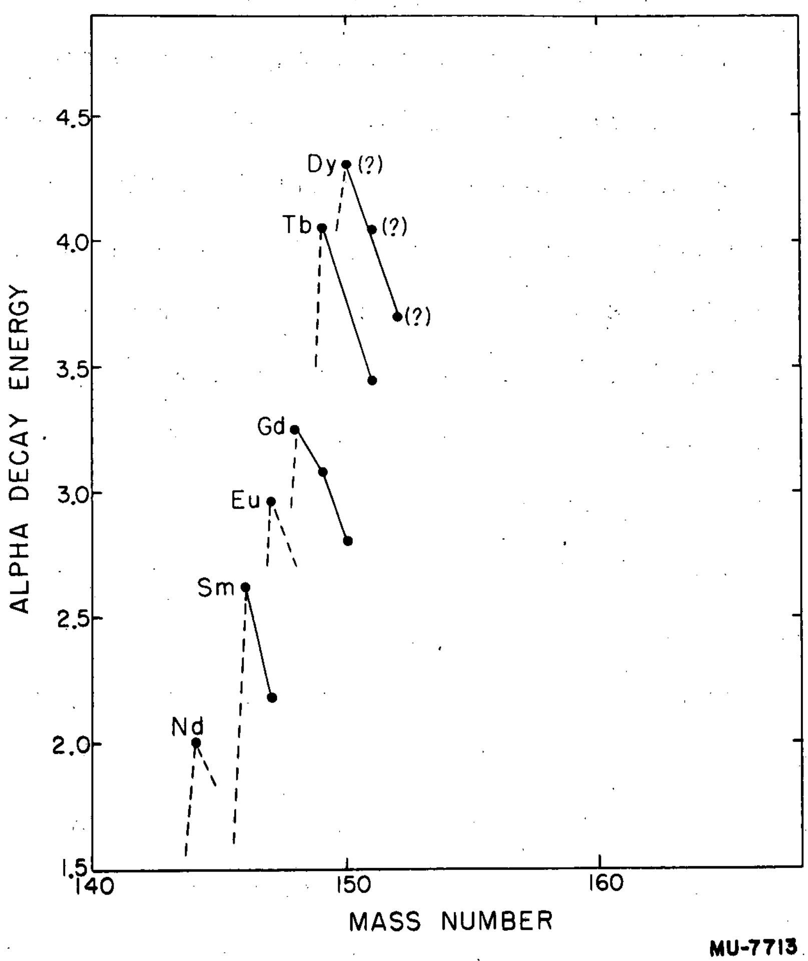

Fig. 5. Alpha decay energy vs. mass number in the rare earth region.

region, the basic structure as related to the 82 neutron shell is unmistakable. The point assigned to Nd^{144} is of special interest because Nd^{144} is a component of natural neodymium (61).

COMPLEX ALPHA SPECTRA

As in other decay processes, the appearance of multiple groups in the alpha emission process may be considered as the result of competition in populating available energy levels. It will be seen that alpha decay lifetimes are influenced by a number of factors and among these is the sharp dependence of lifetime with decay energy. Consequently it would not be expected that transitions to high-lying levels (say 1 Mev) would be readily observed. There are, however, selection processes operating which can delay the highest energy group and cause lower energy groups to be the most prominent. As yet there is no systematic formulation of "selection rules" for the alpha decay process. The development of alpha decay theory with this as an objective is undoubtedly the most important step now faced.

The present discussion will be concerned largely with the regularities which are appearing with respect to the location of energy levels, their spectroscopic designation, and in the degree to which these states are populated. As already stated, only a start has been made in developing reasons for preference in populating certain states.

Even-Even Alpha Emitters

Principal alpha groups (the ground state and first excited state). -- With a high degree of certainty it can be said that the transition to the ground state is the most abundant for this nuclear type. In each of the many cases which have been examined in detail, the main alpha group is that of highest energy and there is no evidence that the group is followed by gamma radiation. To this extent at least the even-even alpha emitters are well behaved according to existing alpha decay theory.

In examining a number of even-even alpha emitters around uranium and in the transuranium region, it was noted that each had a second prominent alpha group of some 40-50 kev lower energy than the ground state transition (62). The abundance of this group was always in the range 15 - 30 per cent.[6] (We shall see presently that the states reached by these alpha groups apparently all have spin 2 and even parity and we shall call each the <u>first even spin state</u> or simply the <u>first even state</u>.)[7] When lower elements or lower neutron numbers of a particular element are considered, the same states are identified but the energy level spacings above the ground states become progressively greater and the abundances of the alpha groups populating these states progressively lower. A summary of the energy spacings between the ground state and first even state as a function of neutron number and proton number is shown in Fig. 6. The points divide into families according to atomic number and appear to reach maxima for nuclei with 126 neutrons. In some cases, the points in Fig. 6 were determined from gamma ray spectra rather than from observation of the alpha groups.

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[6]We shall define "abundance" to indicate the number of alpha particles in a particular group relative to the total number of alpha particles.

[7]It is worth recalling that in conserving parity in the alpha decay of an even-even emitter, the even spin states must have even parity and the odd spin states odd parity. A corollary is that gamma ray transitions from any of these excited states to the ground state must be <u>electric</u> transitions and not <u>magnetic</u>.

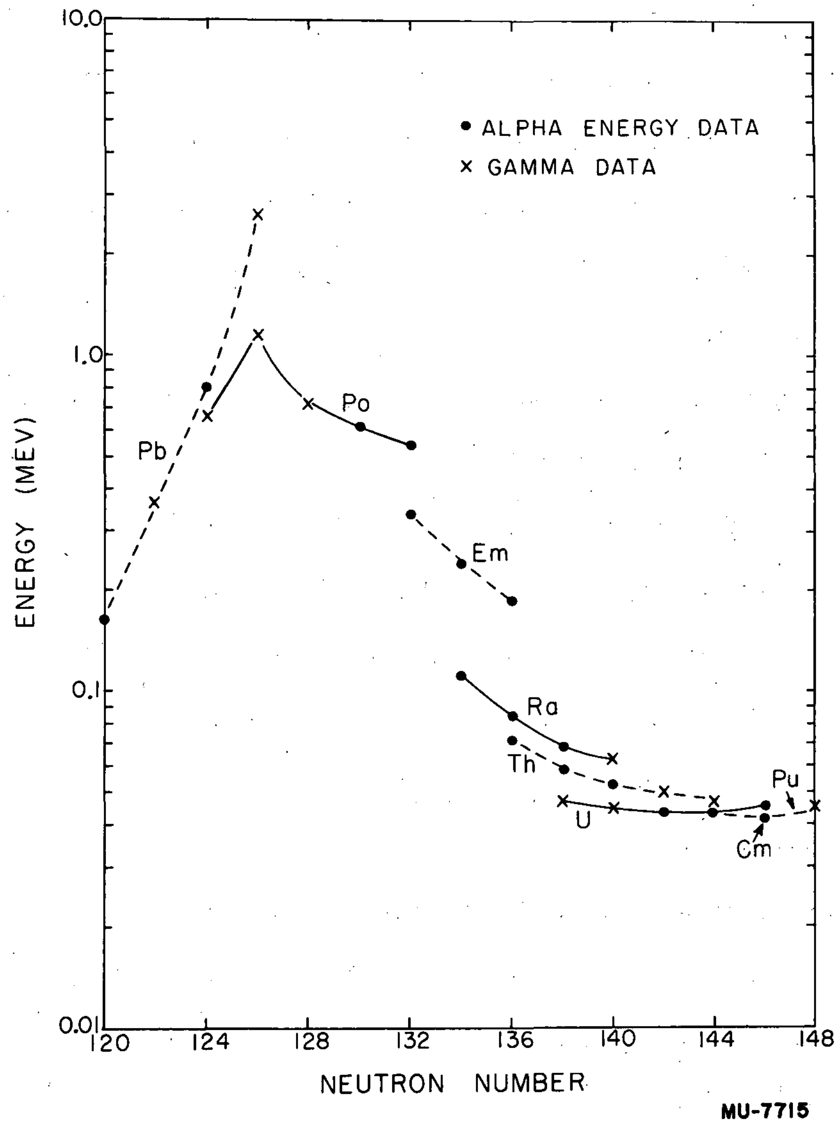

Fig. 6. First excited state energies of even-even nuclei in the heavy element region.

It has already been deduced by several authors (63, 64, 65) that the first excited states of even-even nuclei in general are 2+ states and this applies to regions other than that of the heavy elements and irrespective of the particular energy spacing of the levels. There is theoretical justification for the 2+ assignment both from modifications of the independent particle model (66, 67) and from the collective model (68, 69). There is also theoretical explanation (67, 69) and empirical proof (65, 70, 62, 71) that the energy level spacing for the first excited state goes through a maximum at each closed shell as is seen in Fig. 6 for the 126 neutron shell.

Two typical even-even alpha spectra are shown in Fig. 7 and for the present we shall focus attention on the two lowest states corresponding to the two alpha groups considered. The states assigned 2+ are the first even states already mentioned and are designated so because they de-excite to the ground state by electric quadrupole (E2) transitions.[8] Since there is no evidence to the contrary we shall assume that the first even state is in each case the first excited state.

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[8]The nature of the transition has been deduced from measurements of absolute conversion coefficients, relative L subshell conversion coefficients and α-γ angular correlations. The cases studied are the alpha emitters Po^{210} (72), Ra^{222} (7), Ra^{224} (73, 74, 75), Ra^{226} (75), Th^{226} (7), Th^{228} (9, 76, 77, 78), Th^{230} (77, 79), U^{230} (7), U^{232} (8), U^{234} (8), U^{236} (80), Pu^{238} (13), Cm^{242} (15) and Cf^{246} (89). The excited states in question belong, of course, to the alpha decay products.

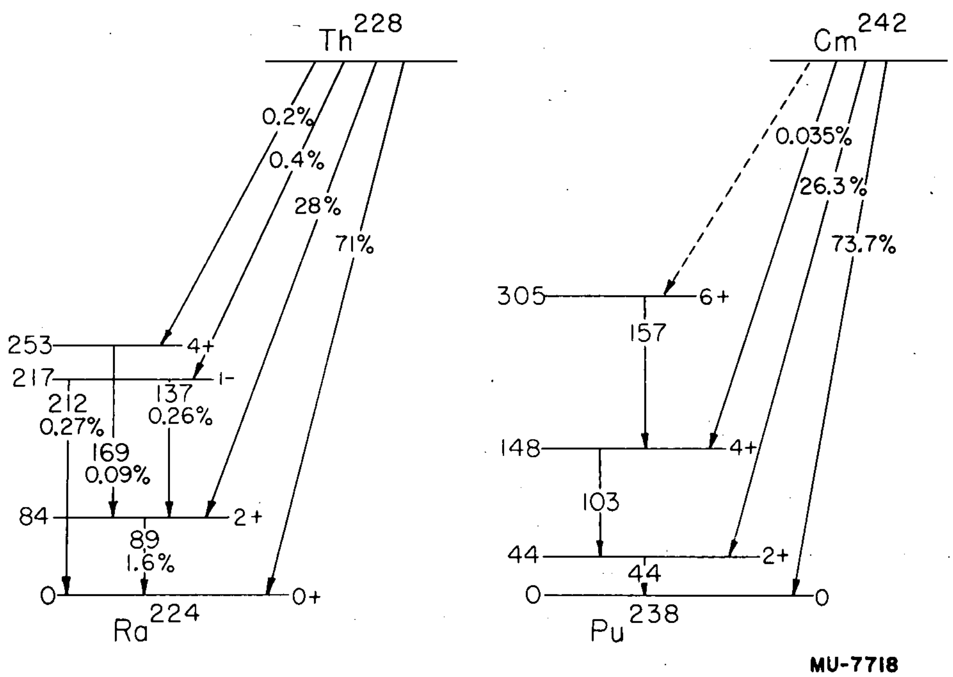

Fig. 7. Decay schemes of Th^{228} and Cm^{242}.

Rare alpha groups (higher even states and first odd state). -- Many of the alpha emitters which have lent themselves to detailed analysis have proved to have one or more additional groups of lower energy and in low intensity. The low intensities of these groups are not attributable alone to lower energy than the two main groups. If we calculate the lifetimes for the ground state transition and that to the first excited state according to current alpha decay theory, the results are in substantial agreement with the measured lifetimes. However, the measured partial half-lives for the higher transitions are considerably longer than the calculated values and we refer to such transitions as hindered. This subject will be dealt with under the section on alpha decay theory and for the present we will consider only the energy level spacings and spectroscopic assignments.

In each case which could be examined in the necessary detail, there was found a rare alpha group going to a state which decays by an E2 transition only to the 2+ state.[9] These states are those designated as 4+ in Fig. 7 and will be known as the second even states. From the nature of the gamma ray transition, the second state could be 0+, 2+ or 4+; the 4+ assignment is made largely from a theory to be mentioned presently which fixes it as a member of a rotational band of states 0+, 2+, 4+ ······. The fact that no crossover transition is seen from the second even state to the ground state may also be taken as partial evidence for the assignment.

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[9] Conversion coefficient data defining the transition as E2 have been obtained in the alpha decay of Cm^{242} (15), Pu^{238} (13, 81), U^{232} (8), Th^{228} (9). For other cases there is no direct information, but other regularities to be discussed presently make it highly likely that comparable states are being considered.

In a few cases, very rare gamma rays have been seen (the alpha groups would be below the limits for detection) and are assigned to transitions between the third and second even states (15, 13). In the case of Pu^{238} decay (13), the gamma ray was shown to be in coincidence with that between the 4+ and 2+ states. Since the energy of the state defined by the gamma ray corresponds closely with expectations if it were the third even state (6+) of the rotational band, it has been so designated (see Cm^{242} spectrum, Fig. 7). The theory behind these assignments will now be examined.

According to a theory of Bohr and Mottelson (82, 68), collective aspects of nuclear motion and individual particle aspects are coupled and in a region well removed from a closed shell there should be a series of energy levels corresponding to a rotational band in which only even states (0+, 2+, 4+ · · · · ·) appear for an even-even nucleus. On this basis, electric quadrupole transitions should predominate. Another requirement of the rotational spectrum in this region is that the states lie at energies proportional to $I_j(I_j + 1)$, where I_j is the spin of the jth even state. For the levels 0+, 2+, 4+, 6+, the ratios of 4+/2+ and 6+/2+ would therefore be respectively 3.3 and 7.0. Figure 8 shows an extensive set of these ratios of the first and second even states and it is seen that the agreement with expectations is excellent for the higher neutron numbers but gradually departs as the region of 126 neutrons is approached. Apparently the same behavior can be noted for even-even nuclei in the rare earth region in reference to the 82 neutron shell (83). The point for Pb^{208} in Fig. 8 is of questionable significance since it is not at all certain that the first two excited states as deduced from Tl^{208}

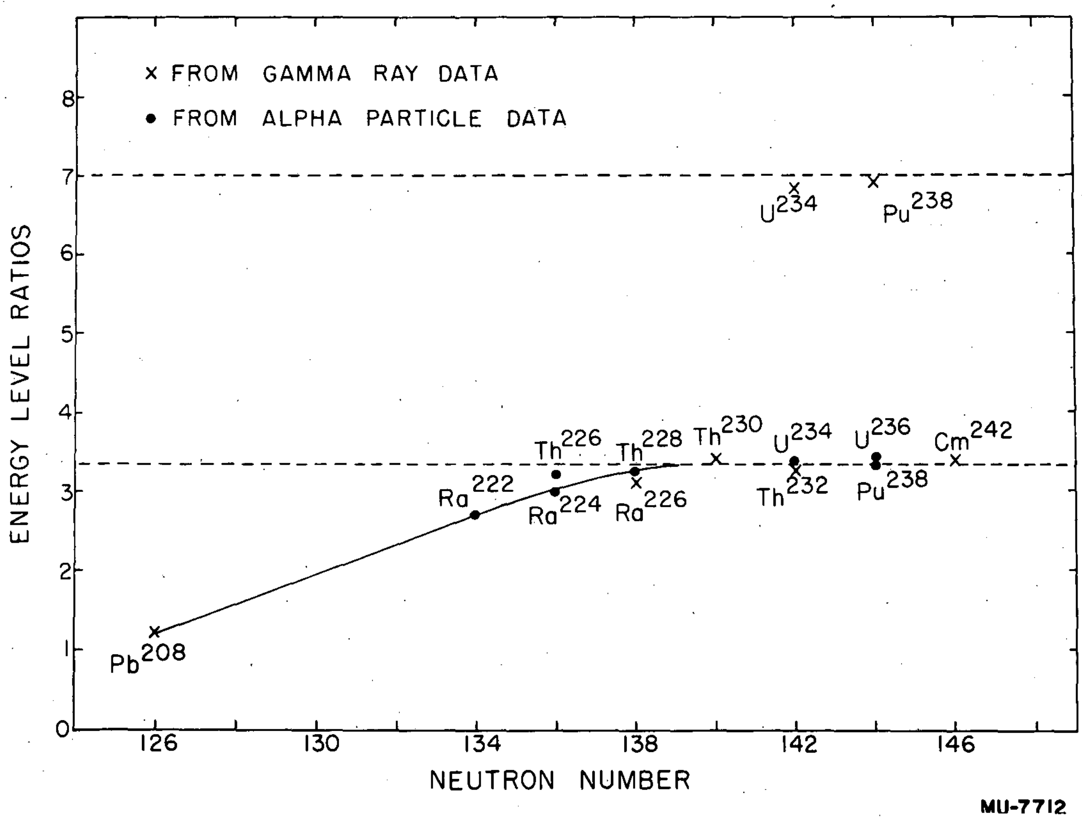

Fig. 8. Ratios of energies of the second and third even spin state to the first excited state. The broken lines represent the expected values from the Bohr-Mottelson theory for rotational levels in regions removed from closed shells.

β^--decay are indeed 2+ and 4+. As an example of this uncertainty, a number of different measurements concerning the first excited state have resulted in different assignments: 1+ or 2+ (84), 2+ (85, 86, 87), 3- (88). In two cases, Pu^{238} and U^{234}, (alpha decay of Cm^{242} and Pu^{238}) where a third even state is inferred, the energy ratio of this level to the 2+ level is indeed close to 7.0. The evidence for the higher state in the case of Pu^{238} decay is that the gamma ray used to define it is in coincidence with the gamma ray from the 4+ to the 2+ states (13).

In a number of cases, a state believed to be 1- has entered among the low-lying even states. The spectrum for Th^{228} which is typical of this type is shown in Fig. 7 (9). In contrast to the second even state, this state always decays both to the first even state and to the ground state. The conversion coefficients of both conform with El transitions (9) as do more recent α-γ angular correlations made on Th^{226}, Th^{228} and U^{230} (89).

The 1- state has probably been identified in the decay of Th^{230} and U^{232} as well as for the three cases just mentioned. Significantly this state has been identified only among the low-lying excited states of nuclei of neutron numbers in the range 134-138 neutrons. From the fragmentary evidence at hand it seems possible that the state has a minimum energy at 136 neutrons and rises at both lower and higher neutron numbers.

With respect to the degree of population of the 1- state in the alpha decay process, the data are too few to arrive at any generalizations. In the cases studied, the process seems to be competitive with that leading to the 4+ state for comparable energies.

Finally it should be remarked that there is no ready explanation for a low-lying odd parity state such as this. Neither the independent particle model nor the collective model would predict such states in any straightforward manner.

Odd Nucleon Alpha Emitters

The alpha spectra of nuclei having odd nucleons are in general considerably more complex than those of even-even nuclei and consequently have not been worked out with the same degree of certainty. Also they may differ from each other considerably and have not yet yielded to a comprehensive generalized picture such as applies to the even-even nuclei.

Nevertheless, some regularities can be discerned, and among these is the appearance of rotational bands (90, 14) closely similar to those of the even-even-nuclei. Interestingly enough, the fundamental state of the rotational band (where such a band shows up distinctly) is that populated in highest abundance, but it need not be the ground state and perhaps usually is not. The important implication of such spectra with respect to alpha decay theory is that powerful "selection rules" are in force since the highest energy transition (that to the ground state) is often in very low intensity (91).

Figure 9 shows some spectra which were selected to illustrate some of the regularities noted.[10] (Obviously it is not possible to discuss all spectra of these types and some, such as that for Th^{227} with eleven reported groups (92), are too complex for analysis at present.)

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[10]The references for the data comprising these alpha spectra are Am^{241} (14, 91), Am^{243} (14), Cm^{243} (15, 8), U^{233} (8).

The partial spectrum for U^{233} is not representative of most of those examined in that it looks much like that of an even-even alpha emitter. The energy level spacings for these states are much like those of Cm^{242} (see Fig. 7) and the relative populations of the states are roughly comparable, but an important difference to be noted is that the second excited state decays both to the first excited state and directly to the ground state. A possible explanation of this spectrum lies in its interpretation as a rotational band, the grounds for which are now sketched.

According to Bohr and Mottelson (68), the rotational band for a nucleus with an odd nucleon will have a fundamental state of spin I_o and higher states of spins I_o+1, I_o+2, $\cdots\cdot\cdot$, all with the same parity. The spacings of these levels will go as $W + CI(I+1)$, where W and C are constants for a particular level sequence. For the U^{233} case the following equations may be set up:

$$E_{98.9} - E_{42.8} = C(I_o+2)(I_o+3) - C(I_o+1)(I_o+2) = 56.1$$

$$2C(I_o+2) = 56.1$$

$$E_{42.8} - E_o = C(I_o+1)(I_o+2) - CI_o(I_o+1) = 42.8$$

$$2C(I_o+1) = 42.8$$

The solution of the simultaneous equations gives $I_o = 2.2$ and since the spin must be half integral we take the closest value, $I_o = 5/2$. It should be mentioned that this analysis is extremely sensitive to the accuracy of the data; an error of only 0.5 kev in the 42.8 kev level, making it 43.3 kev, would change the calculated value of I_o to exactly 2.5. If we accept 5/2 for the fundamental state, the spins of the other two levels become 7/2 and 9/2. The cascading gamma ray transitions could then be M1 and/or E2 and the crossover transition E2, which assignments are not out of order with the fact that both crossover and cascade gamma rays are observed.

The spectra for Am^{241}, Am^{243}, and Cm^{243} alpha decay show quite similar rotational bands with calculated fundamental state spins of 5/2, but the fundamental states in these cases are not the ground states. Americium 241 and Am^{243} differ only in that this state is respectively 60 and 75 kev above the ground state. For Cm^{243}, the ground state transition has not yet been observed even though it has 277 kev higher decay energy than the main group, a fact established by α-γ coincidence counting (15).

It is perhaps not profitable at present to speculate in detail on the meaning of the complex spectra of the nuclear types under discussion. We believe that rotational bands which can be explained as a consequence of collective modes of nuclear motion are discernible. It is also probable that other important features of the spectra must bring in the concept of single particle states. Just what these states are and what guides the alpha decay process to some and not others is still obscure. Some speculations on these matters as they apply to alpha decay theory will be brought out in the section on this subject. It is worth reiterating here that no simple criterion or rule is likely to explain the spectra shown in Fig. 9 as well as the extremely complex spectrum of Th^{227} and that of Pu^{239} which is a good deal simpler but different from both types.

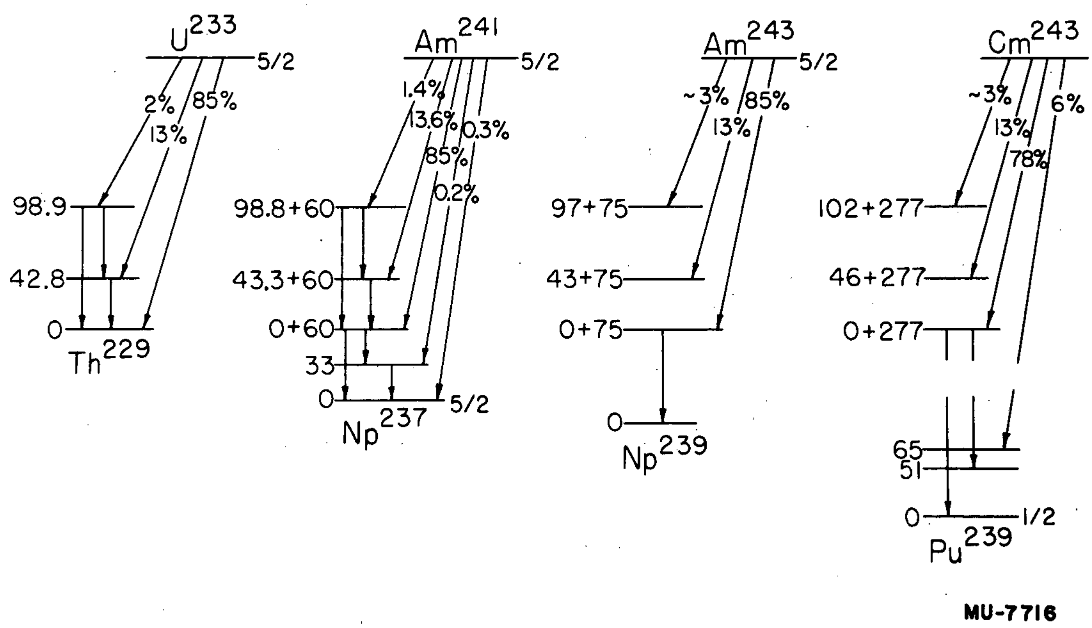

Fig. 9. Decay schemes of some odd-nucleon alpha emitters.

Alpha Decay Lifetimes and Theory

It is possible to correlate alpha decay lifetimes empirically and to arrive at systems which can be used to predict half-lives. For example, regularities can be observed in plotting the half-life with the mass number for successive elements. However, it is also possible to systematize the half-lives in terms of parameters which are involved in current alpha decay theory and thereby also obtain some information on the status of the theory.

It will be seen that the ground state transitions of even-even alpha emitters are treated with extraordinary precision by basic one-body alpha decay theory. However, analysis of the many alpha spectra obtained during the past few years has shown clearly that more elaborate theory is required to account for the behavior of other nuclear types as well as for certain of the transitions to excited states of the even-even nuclei. The development of new approaches to cover these cases is currently in a formative stage. It will not be possible in this review to cover in detail either the old theory or the new approaches; rather, we shall emphasize those aspects of alpha spectra which appear to require new forms for expression and indicate the direction some of these new forms are taking.

Basis for one body theory. -- The initial conception by Gamow (1) and by Condon and Gurney (2) of the alpha decay process as a barrier penetration problem has been cast into a number of forms differing in detail but basically all end up with the decay rate expressed as the product of two factors: 1) a "frequency factor" which may be considered as the decay rate without the barrier, and 2) a "penetration factor" which expresses the probability of an alpha particle emerging through the negative energy region of the coulombic potential barrier. The various solutions to the problem differ in the degree of rigor which is applied and it might be stated that the sensitive portion is the "penetration factor" which is exponential in form and is handled virtually identically in the several treatments. (See, for example, references 93 - 97.)

The parameters which enter into the penetration factor are those which define the barrier for the alpha particle of energy E and these are the nuclear charge and the effective nuclear radius. The frequency factor likewise is a function of the energy of the alpha particle and nuclear radius and although approximations are introduced in its evaluation, this factor is relatively insensitive and does not strongly influence the results. It is probably here, however, that the one body model breaks down as a representation of decay processes, that is, where single particle states are of importance.

It will be noted that of these parameters the nuclear charge has explicit meaning dissociated from the alpha decay process and the alpha energy and decay constant are subject to precise measurement, but the effective nuclear radius is a quantity which is not obviously meaningful for this process when determined from other nuclear phenomena nor do such independent methods of measuring the radius give the desired precision for testing alpha decay theory. The procedure to test the theory is therefore confined at present to see if calculated nuclear radii fall into a reasonable range and pattern of values or fluctuate in an unreasonable fashion (93 - 100, 58). Conversely, one may assume some function for nuclear radii, from this derive theoretical curves to express the decay constant in terms of the other parameters, and test the agreement with the measured decay constants (101, 102, 98, 31, 99). It is this second approach which has proved most useful in correlating a large number of data. Further discussion of this matter will follow as the data for even-even alpha emitters are examined.

Even-even alpha emitters - ground state transitions. -- Figure 10 shows a plot of the half-life versus energy relationship as a family of curves. The curves are defined by the experimental half-lives and are in this respect empirical. If, however, we were to calculate half-lives with the Kaplan (99) modification of the Preston (95) formula by using the measured alpha energy for each point and assuming a function for the nuclear radius, $1.52 \times 10^{-13} A^{1/3}$, the resulting curves would lie close to those of Fig. 10. In detail, the calculated curves of the lower elements (polonium, emanation) would lie somewhat below the corresponding curves of Fig. 10 and for the higher elements (curium, californium) the calculated curves would lie slightly above those shown in Fig. 10. (The segments of curves shown for Po^{210} and Po^{208} and for Em^{210} and Em^{208} should be considered as special cases in which effective nuclear radii suffer a discontinuity and these are nuclei with 126 neutrons and fewer.) To bring about more exact agreement one can bring in a second order effect comprised of the assumption that the nuclear radius parameter should not be a constant, 1.52, but should vary in some fashion, say with atomic number. To see what is required, a calculation was made for the effective radius of each nucleus using measured values of both energy and half-life and the results are shown in Fig. 11. The sharp break at 126 neutrons is apparent, but in addition there seems to be general decrease in radius parameter with increase

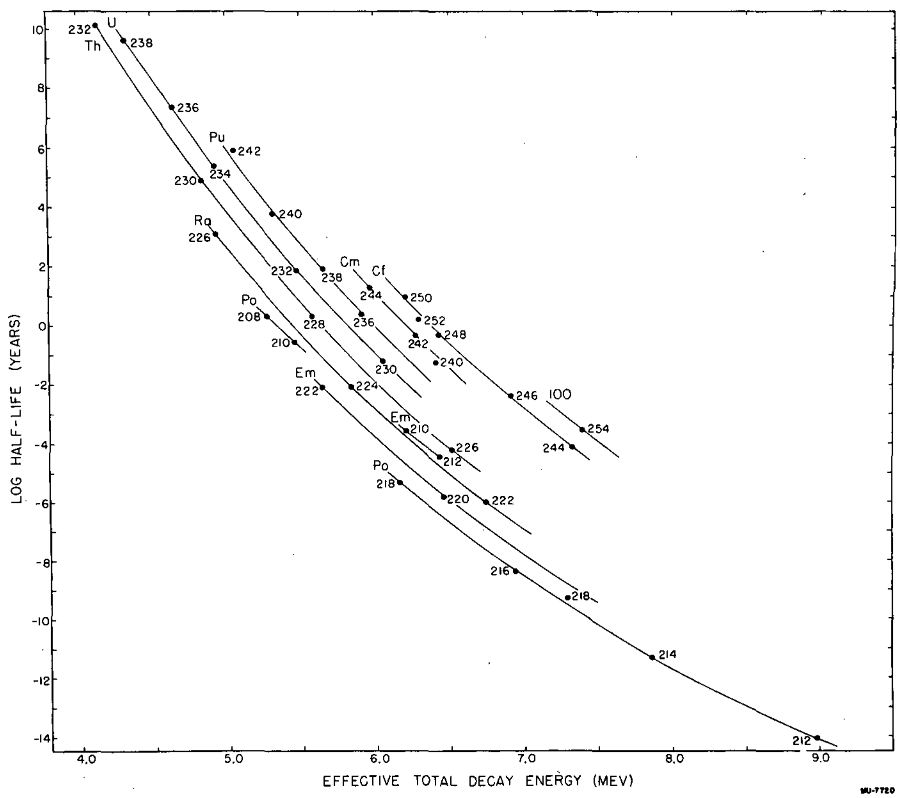

Fig. 10. Experimental values of log half-life vs. effective alpha energy. ("Effective alpha energy" includes correction of particle energy for recoil and electron screening - see text.)

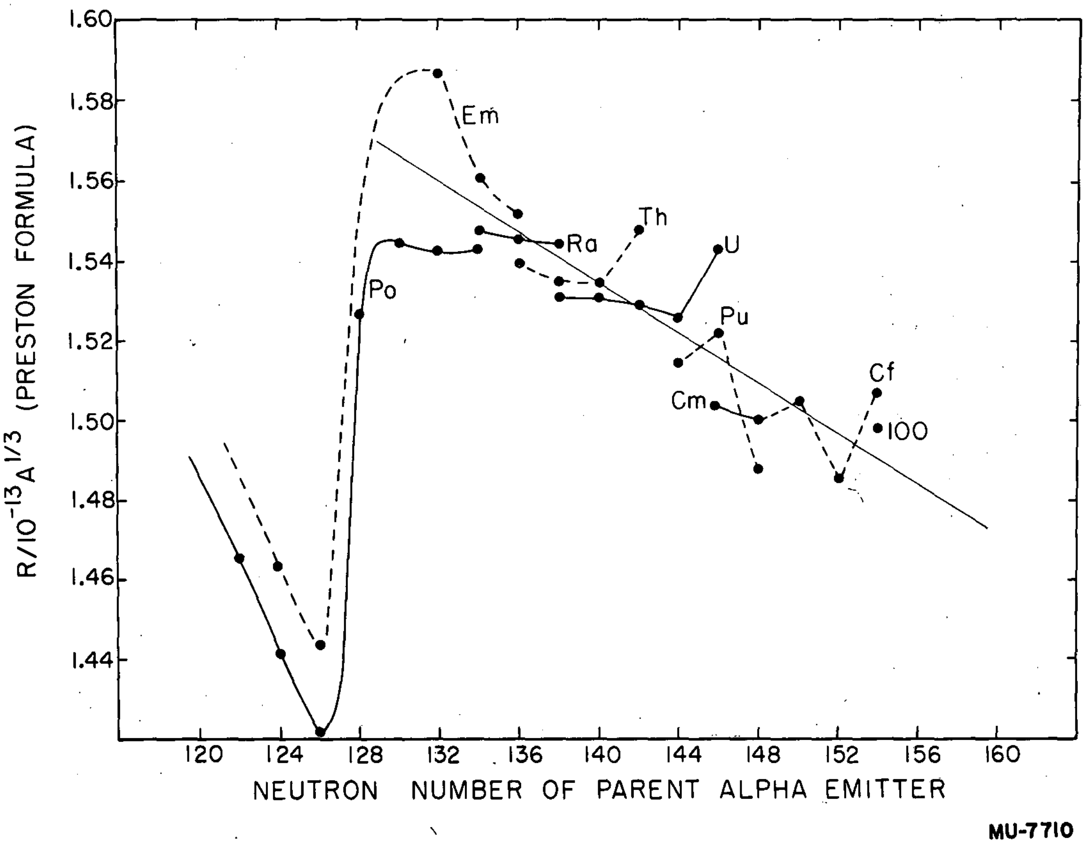

Fig. 11. Nuclear radius parameter.

in atomic number. This, of course, is a restatement of the relation mentioned between the empirical curves and those calculated on the basis of a single average value for the radius parameter. With reference to the data of Fig. 11, it should be pointed out that there is sufficient experimental uncertainty in some of the values which went into these calculations to shift the points by 0.02 units of radius parameter or even more.

In summary we can say that the basic one body theory of alpha decay applied to the ground state transitions of even-even alpha emitters gives a remarkably consistent picture. By using reasonable and consistent assumptions for the values of the nuclear radii, the theory explains observed half-lives which differ by a factor of 10^{24}. It should be pointed out that different formulations of the theory will give somewhat different "best values" for the radius parameter, but each is internally consistent.

Two further points should be mentioned with respect to the curves of Fig. 10. It may be noted that the energy values listed are somewhat higher than measured particle energies. The alpha energy pertinent to alpha decay theory is based on its center of mass velocity and therefore the particle energy must be increased by the energy of the recoil nucleus. This correction amounts to ~100 kev for a 6 Mev alpha particle in this region.

An additive correction to the alpha particle energy so far overlooked has been suggested by Ambrosino and Piatier (103) and has to do with the lowering of the potential barrier by the electron cloud. As discussed by Rasmussen (104), this correction should be applied as an added alpha energy amounting to about the difference of total electron binding energies between parent and daughter in the particular alpha decay process. This amounts to ~38 kev in the region of uranium and is smaller for lighter elements. This latter "screening correction" to the particle energies does not have a profound effect on the agreement of theory and experiment which is under discussion, but it does have the effect of giving somewhat smaller values of nuclear radii than if it were ignored.

The other item concerning Fig. 10 has to do with the reason that smooth curves can be drawn at all because, in principle, each curve is not continuous if the nuclear radius is an independent variable. However, as seen in Fig. 3, alpha energies vary more or less monotonically with mass number A and since nuclear radii do so likewise, each value of energy (for a particular element) does localize the mass number and therefore the radius. Where two isotopes within the same energy range do have large differences in effective radii they do not lie on the same curve. This effect is seen for polonium and emanation curves on the two sides of 126 neutrons.

Even-even alpha emitters - transitions to excited states. -- For any particular ground state transition, one can calculate the partial half-life to any excited state under the assumption that the only factor influencing the relative decay rates is the energy function. Let us first consider transitions to the first excited state (2+). The effective radius as determined from the ground state transition process is adopted and it is assumed that the spin change, whether $\Delta I = 0$ or $\Delta I = 2$, will not affect the decay rates very much. This latter assumption has the theoretical justification of the calculations of Preston (95, 96) which show that contrary to previous treatments, a $\Delta I = 2$ transition should be some 1.5 times faster than a $\Delta I = 0$ transition and that only when $\Delta I = 4$ does the function take the direction of delaying the transition.

The ratios of expected abundances (solely from energy difference) to experimental abundances are shown in Table 2. Around radium, thorium, and uranium the ratios are seen to be close to unity and perhaps to rise significantly for the heavier elements. Some recent ideas on the population of these states will be mentioned below where transitions to the 4+ state are discussed.

One apparent anomaly is Po^{208}, for which no alpha group has yet been seen leading to the 374 kev 2+ state, and the partial half-life is at least 32 fold greater than that calculated. It is significant that there is other evidence (68, 105) that the first excited state of Pb^{204} is not a rotational state. Here, it seems, is a presently isolated but important item which must be included in a comprehensive alpha decay theory. If we may generalize from this single case, we may postulate that a 2+ state of an even-even nucleus is populated "normally" only if it is a collective state of the type discussed.

The examination of transitions to the 4+ state gives a totally different and unique picture (106). Here, if the partial half-lives are calculated as above, it is found that they are much shorter than the measured values; that is, the 4+ states are populated much more sparsely than would be expected on the basis of alpha energy alone. Furthermore, the ratio of measured half-life to calculated half-life varies considerably and in a more or less regular way with atomic number as shown in Fig. 12. It is seen that for Cm^{242} the second even state is hindered in its population by a factor of 400 (compare with factor 1.7 for first even state, Table 2), while for thorium isotopes this factor is of the order of 10. There is no a priori justification for

Table 2

Population of First Even State (2+)
By Alpha Decay of Even-Even Nuclei

Alpha emitter	Energy level (kev)*	Departure factor†	Alpha emitter	Energy level (kev)*	Departure factor†
Po^{206}	163	3.1	U^{234}	52	1.2
Po^{208}	374	>32	‡U^{236}	50	1.1
‡Po^{210}	800	1.6	‡U^{238}	45	1.5
Ra^{224}	240	1.2	‡Pu^{234}	45	3.5
Ra^{226}	188	0.9	‡Pu^{236}	47	2.2
Th^{226}	110	1.2	Pu^{238}	43	1.6
Th^{228}	84	0.9	Pu^{240}	44	1.7
Th^{230}	68	1.0	‡Pu^{242}	45	2.2
‡Th^{232}	65	0.8	Cm^{242}	44	1.7
U^{230}	70	1.1	Cm^{244}	43	1.8
U^{232}	58	1.1	Cf^{246}	42	2.7

*Energy level of the 2+ state of the daughter nucleus.

†Ratio of calculated abundance (solely from energy difference) to experimental abundance. A factor of unity means that the lifetime for the decay to this state (in this case 2+) is according to expectations from alpha decay theory using the ground state transition to define the nuclear radius and neglecting any effect of spin change.

‡The experimental abundances or energies may be suspect.

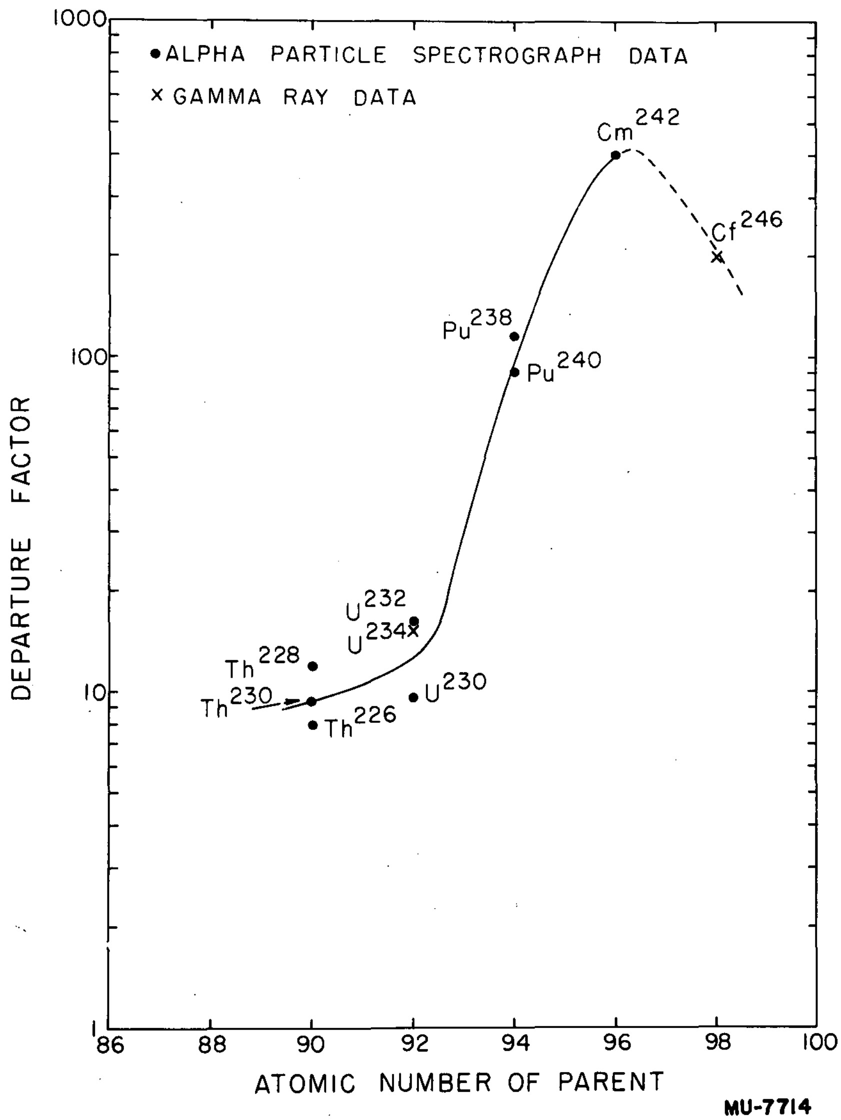

Fig. 12. Departure factors for alpha groups to the second even-spin state of even-even nuclides.

plotting the data of Fig. 12 in the manner shown; for the present it merely serves the purpose of giving visual expression to the considerable range encountered in the population of apparently identical spectroscopic states.

Recently an attack has been made on this problem which appears fruitful. The basic idea developed by Preston (107) is that the alpha particle emerging through the coulomb potential region and beyond the range of nuclear forces can couple with a non-central electric field of the nucleus in such a way as to change its energy. Such fields would be associated with spheroidal distortions in nuclei as discussed by Bohr and Mottelson (68) and Hill and Wheeler (108). The distortion would be described by the intrinsic quadrupole moment whith Ford (69) has shown by calculations to be increasing with mass number in the region above lead and would presumably continue until the approach to a new shell is felt.

The mechanism of quadrupole interaction with the emerging alpha particle wave to explain the population of the first two excited states in a rotational band has been explored in detail by Rasmussen (109). For the case of Cm^{242} decay, for which approximate numerical integrations have been carried out (assuming an intrinsic quadrupole moment, Q_o, in Pu^{238} of $+17 \times 10^{-24}$ cm^2), he found that the population of the 2+ state should be relatively unaffected, but that the population of the 4+ state should be strongly depressed in essential agreement with the experimental observations. The assumed value for Q_o seems reasonable for a nucleus in this region in view of the recent measurement of the quadrupole moment of U^{235} as $+8 \times 10^{-24}$ cm^2 (112), which with the spin of 5/2 corresponds to a value of Q_o of $+22 \times 10^{-24}$ cm^2. The quadrupole moment for even-even nuclei cannot, of course, be measured by spectroscopic means, but there is the possibility of obtaining information through coulomb excitation cross sections and gamma ray lifetimes.

Since this approach in explaining certain features of alpha spectra is in an incomplete state of development, it is not worthwhile to speculate on the possible significance of the downturn of the curve of Fig. 12 between Cm^{242} and Cf^{246}, nor to discuss a number of other ramifications of the mechanism.

We shall just recall that other low-lying states in even-even nuclei have been identified, but for which detailed information is lacking. There is the third even state (6+?) which is a member of the well defined rotational band. Also, in a limited region there appear 1- states which are populated roughly to the same extent as the 4+ states in the same region. It is not clear what type of nuclear configuration would give rise to such states.

<u>Odd nucleon alpha emitters.</u> -- The most obvious question concerning this category of alpha emitters is why the ground state transition is often highly hindered and why the hindrance is so irregular. Table 3 shows the data for such alpha emitters and the last column contains the "departure factor" which, as before, is the ratio of the measured partial half-life for a transition to the half-life which would be calculated from one body alpha decay theory. We note that for the four species shown in Fig. 9 the departure factors for the apparent ground state transitions are: U^{233} -- 1.4, Am^{241} -- 1000, Am^{243} -- 700, Cm^{243} -- >26. It is not certain that for each case in Table 3 the ground state transition has been identified and where this should prove to be the case the departure factor would be larger.

Table 3

Population of Ground State by Alpha Decay of Odd-Nucleon Emitters

Alpha emitter	Particle energy*	Abundance†	Departure factor‡
Cf^{249}	6.00	0.10	300
Bk^{245}	6.33	0.18	700
Bk^{243}	6.72	0.30	900
Cm^{243}	6.049	<0.5	>26
Am^{243}	5.341	$\lesssim$0.003	$\gtrsim$1000
Am^{241}	5.535	0.0034	700
Pu^{241}	5.01	<0.11	>30
Pu^{239}	5.150	0.69	3
Np^{237}	4.86	<0.15	>50
U^{235}	4.58	0.10	900
U^{233}	4.823	0.83	1.4
Pa^{231}	5.042	0.11	220
Th^{229}	5.02	0.10	100
Th^{227}	6.030	0.19	130
Ra^{223}	5.730	0.09	50
Fr^{212}	6.409	0.37	10
Em^{219}	6.824	0.69	18
Em^{211}	5.847	0.33	8
At^{211}	5.862	100	3
At^{210}	5.519	0.32	2
Po^{213}	8.336		1.5
Po^{211}	7.434	0.99	100
Po^{209}	4.877		2

*Particle energy of alpha group believed to lead to the ground state. In some instances it is possible that the groups selected are not the ground state transitions in which cases the departure factors would be greater than those shown.

†Abundances are based on observed alpha groups except where the highest energy group is known to be followed by gamma radiation.

‡Defined in Table 2.

The first point to be disposed of is the effect of spin change. As already pointed out in a number of instances, both theoretical and experimental appraisal indicates that the lifetime is relatively insensitive to spin change, per se, certainly within the framework of reasonable spin changes. Conversely, we know that the ground states of Am^{241} and Np^{237} both have spin 5/2 (110, 111) yet the transition between these states is hindered 1000 fold.

Other ideas, not yet formulated quantitatively, appear to be more promising. One suggestion (31) has to do with the breakdown of the one body model in its implication that alpha particles exist as discrete entities within the nucleus and that the probability of alpha emission depends only upon the frequency factor of this preformed alpha particle in encountering the coulomb barrier and upon the rate of barrier penetration. We have seen that for ground state transitions of even-even nuclei the evidence for this extreme simplification indicates that it is substantially correct. For ground state transitions of odd nucleon emitters, however, it is suggested (31) that there can be a considerable time involved in assembling the components of the alpha particle. Such would be the description if the mixture of configurations which constitutes the emitting nucleus contained only as a minor component the configuration with the odd particle as a constituent of an incipient alpha particle. Rasmussen (90) has pointed out further that nuclear spins need not give a true picture of the nucleon states of the parent and daughter nuclei. In a region well away from a closed shell where large spheroidal distortions are expected, the strong coupling configurations may predominate and the nuclear spin can be different from and lower than the j-value for the single odd nucleon. On this basis, the spectroscopic state of the odd proton in Am^{241} may be quite different from that in Np^{237} even though both nuclear spins are 5/2. If such were the case, the change in wave functions could properly introduce a hindrance to the alpha decay. Only for those transitions where the single particle wave function remains unaltered would one expect unhindered alpha decay. Significantly, it appears that such transitions can be found in the spectra of the odd nucleon emitters, but in general these are not the ground state transitions. This subject will now be examined.

In Fig. 9 it is seen that the most abundant groups of Am^{241} and Am^{243} lead to states which are 60 and 75 kev respectively above the ground state. Furthermore, for the particular energies of these alpha transitions, the hindrance or departure factors are only of the order of unity (in contrast to 1000 for the ground state transitions) and in this respect appear to be like the ground state transitions of even-even nuclei. By making certain assumptions it is possible to deduce the spins for these excited states (60 and 75 kev states). The analysis has already been carried out on pages 27 and 28 and is based on the recognition of three excited states as members of a rotational band from which the spins of these states can be deduced. In this case, the spins for the 60 kev excited state of Np^{237} and the 75 kev state for Am^{243} are deduced to be 5/2 (90, 14). It is suggested (90) then that these states may be those in which the odd particle wave function rearrangement is a minimum and that the transition is therefore relatively unhindered.

As already discussed under even-even nuclei, the splitting of alpha groups into a rotational spectrum may involve the effect of electric field interactions. The appearance of what looks like rotational bands in these odd nucleon species implies, of course, that the same mechanism is in play here.

It seems apparent that the past few years have seen a departure from classical alpha decay theory as well as a reinforcement in our confidence in some of its aspects. It also seems likely that some of the new ideas when further developed will add appreciably to the explanation of the alpha decay process and the ramifications will give a better knowledge of nuclear structure.

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