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SYSTEMATICS OF ALPHA-RADIOACTIVITY

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# SYSTEMATICS OF ALPHA-RADIOACTIVITY

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## ABSTRACT

Correlations of alpha-decay energies in terms of mass number and atomic number have been made for all of the alpha-emitting species now numbering over 100. For each element isotopes show increase in alpha-energy with decrease in mass number except in the region of 126 neutrons where there is an explainable reversal. This reversal has the effect of creating a region of relatively low alpha-energy and long half-life at low mass numbers for such elements as astatine, emanation, francium, and possibly higher elements as had been noted already for bismuth and polonium. Methods and examples of using alpha-decay data to define the energy surface in the heavy element region are discussed. The regularities in alpha-decay are used for predictions of nuclear properties including predictions of the beta-stable nuclides among the heavy elements.

The half-life vs. energy correlations show that the even-even nuclides conform well with existing alpha-decay theory, but all nuclear types with odd nucleons show prohibited decay. The reason for this prohibition is not found in spin changes in the alpha-emission but in the assembly of the components of the alpha particle, and this theory is discussed further in terms of observations made on nuclides having two or more alpha-groups. Using most of the even-even nuclei to define "normal nuclear radius" calculations are now able to show the shrinkage in the regions of lead and of 126 neutrons to amount to about 10%. The much greater change in "effective radius" for bismuth isotopes can be dissociated into the effects of odd nucleons superimposed on the actual decrease in nuclear radius. The simple expression $r = 1.48 \ A^{1/3} \cdot 10^{-13}$ cm seems to fit the data for the even-even nuclei outside of the region of 126 neutrons better than more complex functions.

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## INTRODUCTION

Interest in the systematics of alpha-decay properties goes back to the early
studies of the natural radioactivities at which time it became apparent that there exists
a direct relationship between alpha-energy or velocity and decay constant.[1] Since
then there have been numerous efforts to correlate alpha-decay data on an empirical
basis and, most important, the successful application of quantum mechanics to give insight
into the nature of the process.[2]

Even in general terms the importance of correlations of alpha-decay properties
takes several forms. The discovery of artificial radioactivity and the continued develop-
ment of the means for producing unstable nuclei have made it possible to examine the
properties of an increasingly wide variety of nuclear species. In the case of the heavy
elements in which alpha-radioactivity is prevalent it is most important to be able to
predict, even roughly, the radioactive properties of unknown species because of the
extreme range of half-lives that may be encountered. At the present time such predictions
can be made with a fair degree of confidence and this facility has aided markedly
in the preparation and identification of new nuclear species in the heavy element
region. It is perhaps obvious but worth pointing out here that insofar as preparation
and observation of new nuclides are concerned, predictions of beta-stability must be
considered along with alpha-decay properties.

Another major reason for interest in alpha-decay properties is the insight which these
data afford into the nuclear structure of the heavy elements. Alpha-decay energies
can be measured precisely and in general are thought to give unambiguous values for
decay energies. Since the heaviest elements are joined to the region around lead

by continuous decay series it is possible to interrelate on an energy content basis

most nuclear species in this region making use of few serious approximations. In

brief, it is possible to determine regions of greater nuclear stability from neighboring

regions in which the nucleons are less firmly bound. Lastly, so far as this intro-

duction is concerned, it is possible to gain further information on nuclear structure

by considering the decay constants in terms of the factors which determine decay constants

according to the quantum mechanical explanation for the alpha-decay process and to show

to what degree alpha-decay theory is adequate in interpreting all of the new data.

In the last few years the extension of available data has become so consider-

able as to demand a reexamination of regularities in alpha-decay properties. About

10 years ago only 24 alpha-emitters were known and these were all members of the

naturally occurring radioactive families. In the next five years only five more

had been reported, one produced by artificial means and the others discovered by

more careful examination of the radioactivities occurring in nature. At the present

time about 100 alpha-decaying nuclear species have been reported.[3] Most of the

large number of new additions may be classified in groups with regard to position

among the elements and means of formation. The work on transuranium elements at the

University of California and at the wartime Metallurgical Laboratory of the University

of Chicago has resulted in the discovery of 16 alpha-emitters in this region.[4] The

preparation of $U^{233}$ in quantity has added 7 more alpha-emitters as members of the

$4n + 1$ radioactive family[5,6] while 4 and 18 new species, respectively, were added

by the discovery of the $U^{230}$ collateral series[7] and five new short-lived collateral

series.[8] Another group of 18 alpha-active species resulted from preparation of neutron

deficient isotopes of bismuth, polonium, astatine, emanation and francium.[9]

The additional alpha-emitters which have been characterized within the

past few years are most important because they extend the nuclear types for which

alpha-decay is observed and some also fill in gaps between previously known isotopes.
The most important new regions for which data are now available are the transuranium
elements and isotopes lying on the neutron deficient side of beta-stability. The
band of alpha-emitting nuclei has therefore been extended both vertically and laterally.

The regularities in alpha-decay properties are now sufficiently well
defined and are broad enough in their coverage to make possible meaningful pre-
dictions for all nuclides from bismuth to elements beyond curium. Most of the
available alpha-decay energies and some data of half-lives have already been
reported in recent brief communications.[10]

## ALPHA-DECAY ENERGIES

General Trends.-- There are clearly a number of ways in which to correlate
alpha-decay energies. The most definitive presentation would consist of an energy
surface from which could be measured nuclear instability of all types. However,
there are great advantages in terms of simplicity and magnification of scale in
plotting alpha-decay energies directly. In diagrams of this type one arrives at
a family of curves which take different forms depending upon what the alpha-energy
is plotted against and which parameter is chosen as the basis for joining points.

One of the early methods of treating data was that of Fournier[11] who plotted
the alpha-particle velocity vs. mass number and showed that upon joining isotopes
of each element a family of parallel straight lines resulted, a few points being
notable exceptions to the rule. Schintlmeister[12] has made a similar diagram but
in addition has joined points of the same "neutron excess" (A-2Z). It will be recognized
that members of a decay series linked directly by alpha-particle decay will all
have the same "neutron excess". More extensive treatment and interpretation of
available data was made by Berthelot[13] and more recently by Karlik[14] using methods
somewhat similar to those mentioned.

Another method of correlating data on alpha-decay used recently by Wapstra[15] and Gluekauf[16] consists of a numerical indication of mass defect or alpha-decay energy at the position for the appropriate nuclide on a plot of A vs. A-2Z or Z vs. A-Z. Contours drawn through points of equal instability serve to map trends in this property.

The usefulness of any plot of alpha-decay properties may be measured by the accuracy with which properties of previously unobserved alpha-emitters may be predicted. This in turn depends upon how regularly the data fall into the adopted scheme. There is probably little to recommend one method over another for general purposes since similar uncertainties in extrapolation are inherent in all methods and similar deductions of nuclear properties may be made from all.

At the present time there are sufficient data available over a range of mass numbers for almost every element from bismuth to curium that trends in alpha-decay properties show up well on a plot of alpha energy vs. mass number in which isotopes of the elements are joined. Figure 1 shows such a treatment of the data in which the ordinates give the alpha-disintegration energies* for the ground state transitions which would include gamma ray energies for those cases in which the most energetic alpha particle observed is known to be followed by gamma radiations. If we confine our attention to the heaviest elements, neglecting, for the moment, astatine and lower elements, the most apparent characteristic of the alpha-energies is that the isotopes of each element may be joined by a moderately straight line with increase in energy for decrease in mass number. The trend is roughly linear; that it departs from this condition is not surprising and possible reasons will be discussed below.

The general trend of increase in alpha-energy with decrease in mass number probably can be adequately explained for the present by considering sections of the energy surface at constant mass number. Figure 2 shows a series of such sections

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* In this paper the total energy of the transition between ground states will be termed "alpha-disintegration energy" or more simply "alpha-energy" while the kinetic energy of the alpha particle will be explicitly designated "alpha particle energy".

in idealized form for A, A-2, A-4 etc. The parabolic form of these sections at constant A was deduced by Bethe and Bacher[17] from their semi-empirical mass equation and this was transformed into a more convenient form and the parameters evaluated by Bohr and Wheeler.[18] These Bohr-Wheeler parabolas therefore show energy relationships between isobars. The abscissa in Figure 2 is not continuous. This was done in order to spread the parabolas apart and the notation of Z on each indicates the positions of the same atomic number.

It will be recalled that in the region from lead to uranium there are added on the average almost two neutrons for each proton in attaining a comparable configuration with respect to beta-stability for succeeding elements. Therefore, if as shown in Figure 2 element Z lies at the vertex of the parabola for mass number A-2, element Z-2 will lie approximately at the vertex of A-8. It is further assumed that the change in packing fraction is regular in this region so that if the ordinate of Figure 2 is a function of the mass defect or packing fraction the parabolas will lie as shown. It is a property of identical parabolas arranged in this fashion that the vertical projections of lines representing alpha decay drawn between points on two parabolas (e.g., A,Z to A-4,Z-2) will increase as A decreases. As long as sections of the energy surface at constant A are roughly similar parabolas without sharp irregularities and the slope of the surface is fairly constant, decrease in mass number for each element should be accompanied by increase in alpha-energy. Kohman[19] has developed an expression for alpha-energy in terms of the Bohr-Wheeler mass equation[18] from which the same deductions can be made analytically as those developed graphically here by making equivalent assumptions as to the form of the energy surface. It may be mentioned that the section of the energy surface at constant A will be defined by a single parabola only if A is odd but the same reasoning applies for the condition of "even A" in which odd-odd nuclei lie on a parabola of higher energy than even-even nuclei since alpha-decay is a transition between the same nuclear types.

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The above explanation with Figure 2 considers the parabolic sections of the energy surface to be identical and displaced uniformly on the energy axis. Obviously neither assumption conforms to reality and some further deductions on the shape of the surface may be obtained from more detailed consideration of the alpha-decay data. To do this it is helpful to visualize which parts of the energy surface are under consideration. Accordingly, a schematic sketch is presented as Figure 3* in which the vertical axis is some function of the mass such as mass defect and the other coordinates are the atomic number and mass number as indicated. The surface has been idealized in that it shows none of the irregularities due to odd and even relationships but these need not enter into the present considerations of alpha decay because alpha transitions occur between the nuclei of the same type. Some contours of constant Z and constant A have been entered as well as a few reference points. Some sharp irregularities are noted in the lower part of Figure 3 and these will be discussed below. For the present it is worth considering what information may be obtained from alpha-decay data in the region above about mass number 216.

In Figure 3 is shown a line ab which is the contour following the bottom of the valley and is sometimes called the line of beta-stability. One may also visualize contours parallel to this stability line which would connect nuclei of the same degree of beta-instability. From alpha-decay energies it is possible to obtain information on the slope of these contours and therefore of the surface. The alpha-decay energies do not indicate the slopes in the direction of the contours but at an angle skew to this direction somewhat as indicated in Figure 3 by the arrow labelled "a". Bearing in mind this limitation in interpretation it is still possible to observe a definite trend in this region which may be described as a steep slope of the bottom of the valley above mass 216, a tendency toward a plateau between 224 and 236 followed by an increased slope at higher mass numbers. These numbers should not be taken rigidly as there are no sharp breaks in slope and there may be superimposed other effects in a direction not parallel with the valley.

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* We wish to thank Mr. Robert Olson for the preparation of this sketch.

The manner in which these inferences were made may be clarified by reference to Figure 4. Here are plotted the alpha-decay energies as a function of mass number, and nuclei of constant Z are connected (with light lines) as in Figure 1. The contours shown in heavy lines on Figure 4 are intended to map regions of constant degree of beta-instability. For example, the contour XY joins points along the stability line ab in Figure 3, lower contours join points on the Bohr-Wheeler parabolas of Z lower than the most stable value of Z and the contours above XY go through points of higher values of Z than the most stable. The ordinate value, that is the alpha-energy, for each contour is seen to go through a minimum indicating a minimum slope to the energy surface in that region. The reason for the vertical displacement of the contours in Figure 4 may best be visualized by examining the model in Figure 3. Since alpha-decay proceeds in a direction skew to that of the valley the alpha energies of points on the left hand slope of the valley will become progressively greater the further one proceeds up the slope and they will become lower the further one proceeds up the other slope.

One may also examine contours in the direction of alpha decay in which points along an alpha decay sequence are joined. Some of these are shown as dotted lines in Figure 4 and again show minima. Here, however, the minima arise from at least two effects, the first caused by the plateau in the direction of the valley and the second effect is that of crossing from one slope of the valley to the other.

It is not possible to attach to this plateau any precise meaning in terms of nuclear structure in the sense that some sharp irregularities in the region around lead and polonium may be interpreted (see discussion below). The existence of this region has the practical importance of allowing a few nuclei in the region of beta-stability to have sufficiently long alpha half lives to persist in nature. Let us examine what would have happened if the slope of the energy surface were a little steeper. Then $U^{238}$ which is assumed still to be beta-stable might have had a 10- or 20-fold shorter half-life and would have essentially decayed away. Now

$U^{240}$ would have a longer alpha half-life but being beta unstable would decay to $Pu^{240}$ which would again have too short an alpha half-life. It may be stated that the conditions for persisting in nature is that a nuclide be the heaviest beta stable isotope of the element, that is that it lie on the proper side of the valley, and that it lie on this plateau. Apparently only two nuclei fulfill these conditions, $Th^{232}$ and $U^{238}$. The other nucleus with long half-life, $U^{235}$, is found in nature for another reason having to do with an abnormally long half-life for its decay energy. This will become clearer when half-lives are discussed.

Returning to Figure 1 it is seen that the smooth trend in energy vs. mass number between the heaviest and lightest isotopes of each element from emanation to curium no longer applies to elements below emanation and in fact even emanation and francium now have identified isotopes which do not fall in this sequence. Here the trend of increase in alpha-energy with decrease in mass number is apparent for the heaviest isotopes of each element but a point is reached at which there is a sharp break and alpha-energy decreases with decrease in mass number. At still lower mass numbers there is a minimum in the curve and the initial trend is again resumed.

Bismuth isotopes present an interesting case in that alpha-activity disappears over a wide range of mass numbers and then reappears at very low masses. The condition of increase in alpha-energy with decrease in mass number is fulfilled from $Bi^{214}$ to $Bi^{211}$ while $Bi^{210}$ has a much lower alpha energy and $Bi^{209}$ (stable bismuth) has an alpha-energy less than 4 Mev as inferred from the inability to detect its alpha-activity. Several bismuth isotopes in the range $Bi^{202}$-$Bi^{207}$ are known[3,20] but none of these have detectable alpha-activity. However, at still lower mass numbers ($Bi^{201}$ and lower) alpha-activity again appears. The same trend but at higher energies is noted for polonium and astatine and probably for higher elements.

This behavior may be explained by a sharp irregularity in the energy surface of the form indicated in the lower part of the model shown in Figure 3. It may readily

be seen that the Bohr-Wheeler "parabolas" at constant A have a decided depression

included as an irregularity and with this it is possible to explain the observed

inversion in alpha-energies in a manner similar to that used in Figure 2. Another method

that may be used is to consider contours of the energy surface at constant Z. This

has been done in schematic fashion in Figure 5 for Z = 84 and Z = 82. The lengths

of the arrows indicate the alpha energies for the polonium isotopes of the mass

numbers indicated and the sequence showing increase in alpha energy from $Po^{220}$ to

$Po^{212}$ followed by a sharp decrease and gradual increase in progressing to lower mass

numbers are indicated in this fashion. The exact shapes of these contours must not

be taken seriously beyond the point that they will have some such shape if the

irregularity in the energy surface sets in at some line of constant neutron number

as shown in Figure 3. The changes in slope of the curves of Figure 5 are made to

fit the observed alpha-decay data. It will be noted that the contours were drawn

through even-even nuclei only. This was done in order not to complicate the drawing

by odd-even alternations. The meaning of the inversion in alpha energies noted in

this region is attributable to abnormally strong nuclear binding associated with

126 neutrons and will be discussed further below.

The preceding paragraphs have dealt with the explanation of the general

trends in alpha decay energies. It is the purpose here to discuss the data presented

in Figure 1 and to point out some of the deductions which can be reached concerning

the properties of a number of individual nuclides.

Evaluation of Data.-- It is inherent in most of the methods used for determining

alpha-energies that fairly accurate values are obtained. Almost none of the energies

shown should be in error by more than 100 Kev, and most of them are known with considerably

better accuracy. It will be seen in the final section of this paper that there is

hope of making calculations of nuclear radius from alpha decay data in which uncertainties

in alpha energy of the order of only 10 Kev would be meaningful and undesirable. However,

for the present considerations such accuracy is not necessary to observe the general

trends and the principal uncertainties in interpretation arise from other sources.

There are a number of cases of uncertain assignment of mass number and these are

indicated in Figure 1 by superscript question marks following the mass number. In

no cases were the assignments as indicated purely arbitrary; for example, the four

neutron deficient bismuth alpha emitters were arranged in the order shown by excitation

function data and the mass numbers of some were assigned by observing the genetic

relationship with the lead and thallium daughters arising through successive electron

capture decay.[21]

Other entries in Figure 1 are accompanied by question marks preceding the

symbol as in the case of $Pu^{232}$ indicating uncertainty of the energy value, due in this

instance to the lack of resolution of this group from others formed in the same

irradiation. All energies which are calculated or estimated are similarly denoted.

Beyond the uncertainties already mentioned it should be realized that total

decay energy is the property which should be compared and there has been a tacit

assumption that the alpha-energies shown represent ground state transitions. Where this

condition is known not to apply, corrections have been made by addition of the gamma

ray energy as in the cases of $U^{235}$ and $Am^{241}$ (see Appendix for details). There is

good reason to believe that there may be many cases, as yet unproved, in which the

observed alpha particles do not correspond to the ground state transitions. As

examples, $Pu^{239}$ and $U^{233}$, both of which would seem to be out of line in Figure 1, have

associated with them considerable gamma radiation[3] which may mean that the alpha

groups of the ground state transitions have not been observed. Further reasons lending

credence to the existence of this phenomenon will be brought out in the discussion

of half-life vs. energy relationships.

The alpha energies for three of the entries, $Bi^{210}$ (RaE), $Pu^{241}$ and $Am^{242}$,

have been calculated from closed decay cycles since the alpha particles could not be

measured directly. In all three cases the alpha-decay process has been evidenced

by the appearance of the daughter isotope. Broda and Feather[22] identified the 4-min.

thallium as the alpha-decay daughter of 'RaE and using its beta-decay energy, that of

RaE and the alpha-energy of $Po^{210}$, it was possible to calculate the alpha-energy of

RaE. The following diagrams designate the data used in calculating the alpha-energies

for $Pu^{241}$ and $Am^{242}$ (see Appendix for further discussion)
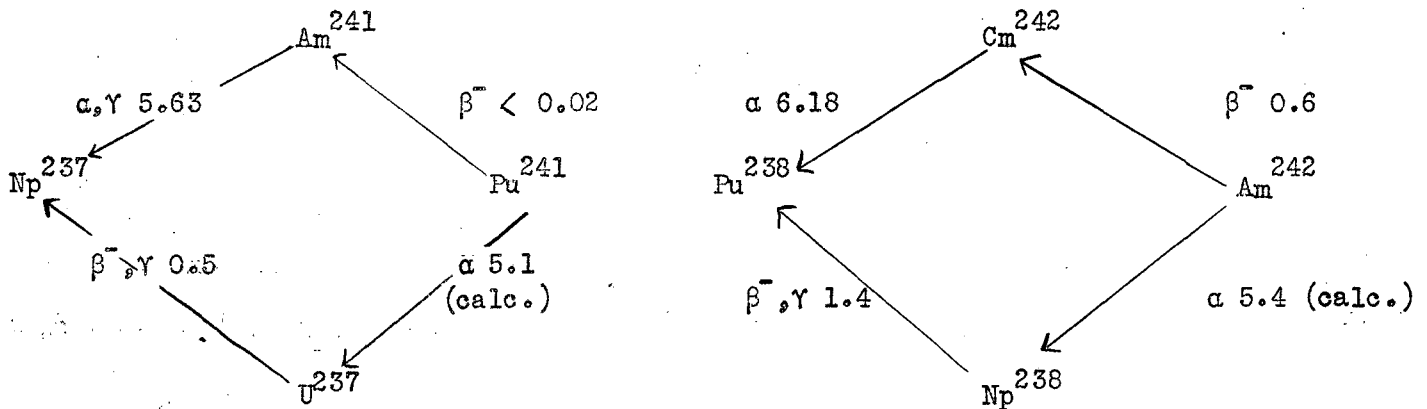


Bismuth and Neighboring Elements, the Effect of 126 Neutrons.-- The recently

discovered alpha-radioactivity of highly neutron deficient bismuth isotopes (Figure 1)

is of interest in marking the "reappearance" of alpha-radioactivity in bismuth at a

considerable departure in mass number from the alpha emitters among the natural

radioactivities. A possible explanation has already been advanced in which the trend

in alpha energies for these light bismuth, polonium, and astatine isotopes is thought

to be a resumption of the trend which all of the heavy isotopes show, namely an increase

in alpha energy with decrease in mass number. This resumption in trend follows an in-

terruption caused by a region of abnormally great nuclear binding.

From the data at hand it is readily seen why no alpha activity has been

noted for previously known isotopes such as $Bi^{206}$ and $Bi^{204}$. By extrapolations of the

curves for bismuth in Figure 1 it may be estimated that these isotopes have alpha-

decay energies around or below 4 Mev. For this energy the alpha half-life would be

perhaps greater than $10^8$ years and therefore alpha particles would be undetectable. As

will be discussed below, it would be illuminating to have available the partial decay

constants for alpha-emission for these electron capturing bismuth isotopes since the relationship between half-life and energy gives further information on nuclear binding through the effect on nuclear radius. The 5.15-Mev alpha-emitter thought to be $Bi^{201}$ has a measured half-life of one hour and even a crude estimate of its alpha-decay half-life would set it at about 50 years illustrating the extremely low alpha-branching.

The reappearance of high alpha-energies which allow alpha-emission to be detected for the highly neutron deficient isotopes of bismuth, polonium, and astatine gives rise to the thought that elements of still lower atomic number might similarly show alpha-activity. This matter has been considered at greater length by Kohman.[19] Recently in this laboratory[23] there has been observed a number of short-lived alpha emitters in this region following irradiations of gold with high energy deuterons and two of these have been tentatively assigned to isotopes of gold and mercury. On the other hand, similar experiments[24] were able to show no alpha-activity in lead. This may mean that the region of stability which causes a decrease in alpha-energy and abnormal increase in half-life in a localized region for astatine, polonium, and bismuth is intensified and broadened at the position of lead and possibly thallium so that alpha-activity is no longer exhibited. When this region is passed alpha-energies and alpha-decay constants are again sufficiently high especially for neutron deficient isotopes.

In considering the reason for the inversion in alpha-energies of bismuth, polonium, and astatine it is necessary to postulate a region of abnormally stable nuclear binding. In proceeding from high toward low mass numbers there is good evidence that this effect sets in at some constant neutron number for each element as indicated in Figure 3. Thus the highest alpha-energies for bismuth and polonium occur for the isotopes $Bi^{211}$ and $Po^{212}$ and for astatine probably at $At^{213}$ (see next section). All of these decay by alpha-emission into nuclei with 126 neutrons and it is probable that at this neutron number there is abnormally stable nuclear structure as compared

with nuclei having greater neutron number.[25,26] It may be worth pointing out that insofar as alpha-decay energies are an index there is no sharp discontinuity in nuclear binding below 126 neutrons as there is above this number.

Neither from Figure 1 nor from the theory is there any quantitative way of telling how many elements beyond astatine this effect of 126 neutrons will make itself felt. In Figure 3 the depression in the energy surface due to the 126 neutron configuration is shown to extend to element 88, radium. It may also be seen that for each successive element, the minimum in alpha-energy caused by the depression should lie at progressively higher alpha-energies. Examining the isotopes of Bi, Po, and At each with 126 neutrons, the alpha-energies are respectively $<$ 4 Mev, 5.3 Mev and 5.9 Mev. Because the differences between successive elements seem to be converging it is difficult to extrapolate to the next nucleus with 126 neutrons, $Em^{212}$, but a reasonable guess is that its alpha-energy will not be more than 6.4 Mev. As will be pointed out the alpha-decay for nuclei with 126 neutrons or less is highly forbidden and from the curves of Figure 9 one might expect a half-life for $Em^{212}$ in the range several minutes to an hour, depending, of course upon its energy. Because of the 126 neutron configuration, $Em^{212}$ is possibly sufficiently stabilized to be beta-stable. As a result one would predict that at least this light isotope of emanation should be observable. In summary, as far as predictions are concerned, there is good reason to believe that the curve for emanation (element 86) in Figure 1 will reach a maximum at $Em^{214}$ and then at lower mass numbers will descend into a region in which the nuclei will have alpha-energies in the range of about 6 Mev.

Very recently in this laboratory,[27,28] attempts have been made to prepare emanation and francium isotopes of low mass number by spallation of thorium with high energy protons. It was found that there are indeed alpha-emitters in the predicted energy region which must be ascribed to nuclei of lower mass number for these elements than any which had been heretofore examined. In particular, two of these activities

have been assigned; a 23-min. period with particles of 6.17 Mev energy, to $Em^{212}$, and

a 19 min. period with 6.25 Mev alpha-particle energy to $Fr^{212}$.

Although this same effect may exist for several higher elements it should

become less pronounced and increasingly difficult to observe since not only will the

alpha half-lives be expected to become shorter for higher elements due to larger alpha-

energies but at constant neutron number the affected nuclides will be progressively more

highly neutron deficient and therefore have short electron capture half-lives.

## PREDICTIONS OF NUCLEAR PROPERTIES

<u>Methods and Examples.</u>--   It is apparent that one may use the correlations

of Figure 1 to predict the alpha-energies of many nuclides just as has been done

previously by a number of workers without the benefit of the comprehensive data now

available as a guide.  In addition, one may use the half-life vs. energy correlations

which are taken up in a later part of this paper to predict alpha half-lives.  There

will be no attempt here to compile a list of predicted nuclear properties of presently

unknown species or modes of decay.  However, there are a number of generalities of some

importance which may be reached through the use of particular data either available or

obtainable by prediction, and some of these will be examined.  One of these, having to

do with relatively long-lived alpha-activities of emanation and francium isotopes of

low mass number, has already been discussed.  A brief recapitulation of the premises

used and methods which are applied may be in order. -

The first consideration is that all nuclides in the region under discussion

are thermodynamically alpha-unstable and almost all which are either found in nature or

artificially prepared would have measurable alpha-activity if alpha-decay were the

only type of instability in force.  However, alpha-decay may not be discernable in

beta-unstable nuclei if the ratio of beta half-life to alpha half-life is extremely

small.  Likewise, a beta-unstable nuclide may not sensibly exhibit its beta-instability

if the alpha half-life is extremely short.

There are a number of reasons for wishing to know whether a nucleus is beta-stable or unstable irrespective of its alpha-decay properties. The beta-stable nuclei in this region are entirely analogous to the stable nuclei lower in the periodic table and their pattern constitutes a source of information on nuclear structure. An example of this will appear in a later section in which is discussed the possibility that astatine (element 85) has no beta-stable isotopes as is apparently the case for two other elements technetium (element 43) and promethium (element 61). Another, quite different, use of the knowledge of beta-stability has to do with the preparation of nuclear species through the beta-decay process. As an example, the validity of the difficult measurements showing that astatine isotopes arise from rare beta-branching of the "A products" (polonium isotopes) of the natural radioactive series must clearly be questioned for these "A products" which can be reasonably proved to be beta-stable. All in all, the question of beta-stability is of great importance and an attempt is made (Table 1) to list the heavy nuclides accordingly.

Beyond predictions of _type_ it is also important to predict _degree_ which, for alpha-emission, is the principal part of this paper. The accurate forecast of alpha half-life along with similar predictions with regard to beta-stability are the require-ments in determining how best to prepare and identify unknown species of interest. Such predictions were invaluable in preparing successively the transuranium elements, the transplutonium elements and are the guiding factors in attempting to prepare still higher elements. Therefore, in addition to alpha-decay predictions, it is often important to estimate beta-decay and electron capture decay energies and through modified Sargent diagrams, the half-lives.

The methods of making these predictions vary in kind and reliability. Alpha-energies may be read off a plot such as Figure 1 by interpolation or extrapolation bearing in mind the sharp changes which occur in the nuclei near closed shell configurations. For example, one may be fairly certain that the alpha-energy of $Em^{221}$ lies between those of $Em^{220}$ and $Em^{222}$ and has an energy of $6.0 \pm 0.1$ Mev; on the other hand, it would

undoubtedly be erroneous to assign $At^{213}$ an alpha-energy between $At^{214}$ and $At^{212}$ since

from other correlations we would infer that $At^{213}$ has a higher energy than $At^{214}$ (as shown

in Figure 1) in an analogous fashion to the pairs $Bi^{211}-Bi^{212}$ and $Po^{212}-Po^{213}$.

With information on some decay energies it is often possible to calculate others

making use of decay cycles the simplest type of which is shown as follows:
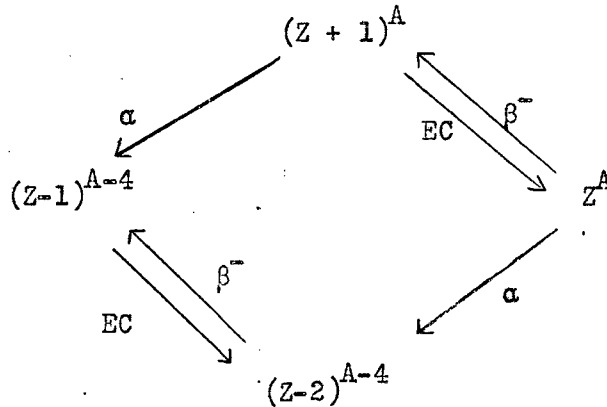


If any three members are known, the fourth is uniquely determined. Some-

times approximations can be made with very little experimental information which in

turn can lead to important deductions. As an example, we may use alpha-decay energies to

deduce something about nuclear spins near closed neutron and proton shells. The case

in point is to obtain information on the spin of $Pb^{209}$ which has one neutron beyond the

"closed shell" 126 and $Po^{209}$ which has one neutron less than 126. Known information

is shown on the following diagrams with broken line arrows indicating paths of decay

not yet observed.
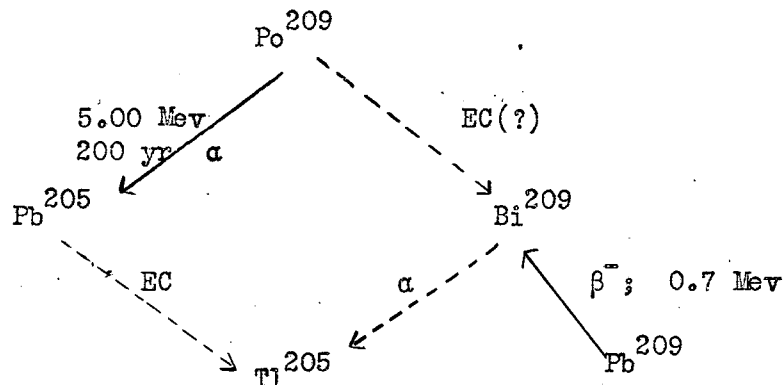
From the failure to observe alpha activity in natural bismuth (minimum half-life
$\sim 10^{12}$ yr.) and making use of the half-life energy relationships considered in a
following part of this paper, it is almost certain that $Bi^{209}$ has an alpha-decay energy
less than 4 Mev and it could be considerably below this, say 3 Mev. Since $Pb^{205}$ must
be heavier than $Tl^{205}$ one can immediately decide that $Po^{209}$ is at least 1 Mev heavier
than $Bi^{209}$ and perhaps considerably heavier. In preparations of $Po^{209}$ showing considerable
alpha-activity the number of x-rays from the electron capture process allow one to set
an upper limit of 10% for the electron capture branching making the minimum half-life
for this mode of decay 2000 years.[24] Realizing that this is a minimum value for the
half-life and that the decay energy is at least 1 Mev and might be considerably
greater, the decay process is seen to be highly forbidden when compared with a large
number of other cases which indicate a half-life of about 100 min. for this decay
energy.[29] The spin of $Bi^{209}$ is known to be 9/2 and since one must postulate a large
spin change in the $Po^{209} - Bi^{209}$ transition, it is necessary to assign a small spin
number such as 1/2 to $Po^{209}$.

The other isobar decaying to $Bi^{209}$ is $Pb^{209}$ of the $U^{233}$ family which has
a 3.3 hr. half-life and 0.7 Mev decay energy. This half-life and energy corresponds to
an allowed transition which Feather and Richardson[30] associate with a spin change
of -1. On this basis one would assign the huge spin number 11/2 to $Pb^{209}$ but in any
case it must be close to that of $Bi^{209}$ and therefore large. The point to be made is
that $Pb^{209}$ with 127 neutrons has one neutron beyond the configuration 126 and according
to Mayer[31] the extra neutron should indeed be in the 7i level with spin term $i_{11/2}$.
Turning to $Po^{209}$ it is seen that it has 125 neutrons, one below the closed shell of
126. According to the term schemes of Mayer, the last levels to be filled in the
shell have the spin term $i_{13/2}$ but as is noted for previous shells these high spin
number levels fill only in pairs and there is crossing with a level of lower spin
number. In this case the 125th neutron would bear the spin term $4p_{1/2}$ which Mayer
lists as the closest level to the first i levels. It is seen that this assignment is

in good agreement with the electron capture decay properties of $Po^{209}$ elaborated above. It might be mentioned that $Pb^{207}$ should be an analogous nucleus (125 neutrons) and Nordheim,[32] Mayer,[31] and Feenberg and Hammack[33] assign the odd neutron to a 4p state with spin 1/2.

For further illustration of the use of alpha-decay data in predicting nuclear properties the cases of two astatine isotopes will be considered.

One of these has to do with the alpha-energy of $At^{213}$ indicated to be 9.2 Mev in Figure 1 although this is only a prediction. It will be noted that $At^{214}$ has almost the same alpha-energy as $ThC'(Po^{212})$ the most energetic alpha-emitter previously known and it would be of interest to know whether $At^{213}$ has still higher decay energy. The point of uncertainty is whether or not $Po^{213}$ is beta-stable and if so what the $At^{213}$ electron capture energy is. Although this cannot be predicted for sure it is probable that it is either $\beta^-$-stable or is only very slightly unstable with respect to $At^{213}$. The beta-stability of $Po^{213}$ is discussed in a following section on beta-stability. Taking the $Po^{213}$-$At^{213}$ beta-decay energy as zero, the total decay energy of the $Pb^{209}$ beta transition as 0.70 Mev and the alpha-energy of $Po^{213}$ as 8.5 Mev, then the alpha-energy of $At^{213}$ is calculated to be 9.2 Mev. This energy would conform with the hypothesis that the highest alpha-energy in this region will reside for each element in that isotope which decays to a product with 126 neutrons, in this case $At^{213} \longrightarrow Bi^{209}$. To this estimated decay energy of $At^{213}$ must be added the electron capture energy of $At^{213}$ or the $\beta^-$-energy of $Po^{213}$ must be subtracted, whichever applies.

$At^{216}$ is reported by Karlik and Bernert[20] to arise in the thorium decay series through $\beta^-$-branching in the decay of $ThA(Po^{216})$. These authors noted a weak 7.57-Mev alpha-group which decayed with the half-life of separated thoron $(Em^{220})$ and was attributed to ~0.01% branching in ThA decay according to the following scheme:

$At^{216}$

$Tn(Em^{220})$
α (55 sec.)

α

$β^{-}$ (0.01%)

$ThB(Po^{212})$

$ThA(Po^{216})$

α (99.99%)

$ThB(Pb^{212})$
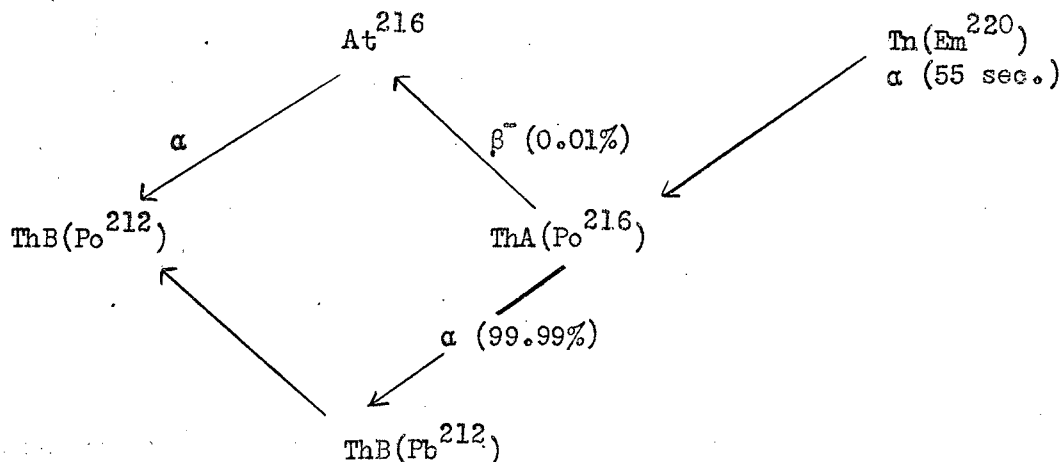
Karlik and Bernert have pointed out a serious difficulty in this interpretation since in summing the decay energies in the closed cycle it turns out that $At^{216}$ (using their value for the alpha-energy of $At^{216}$) is actually unstable with respect to ThA by 0.15 Mev while the apparently observed $β^{-}$-branching of ThA would demand an estimated energy of 1 Mev in the opposite direction, thus introducing a discrepancy of 1.15 Mev. They suggest an explanation retaining their assignment of the 7.57-Mev alpha-group to $At^{216}$ by assuming that ThB decays only to an 1.15-Mev excited state of $Bi^{212}$(ThC) while the alpha-decay of $At^{216}$ proceeds to the ground state. This explanation seems untenable for a number of reasons. If ThC represents the postulated excited state of $Bi^{212}$ then the alpha-energy of the ground state would be 1.15 Mev lower than the measured value (6.1 Mev), that is, about 5.0 Mev. From Figure 1 it is seen that whereas 6.1 Mev falls directly in line between the energies for $Bi^{211}$ and $Bi^{213}$ a value of 5.0 Mev would be completely out of line. In addition, the explanation is in conflict with the known properties of the collateral decay series[8] starting with 22-hr. $Pa^{228}$ because in this series $At^{216}$ arising from the alpha-decay of $Fr^{220}$ decays to $Bi^{212}$ identical in properties with ThC, that is, the 6.1-Mev energy is noted which would not be possible if $At^{216}$ must decay to the ground state of an isotope for which ThC is an excited state. That the entire $Pa^{228}$ decay series has been wrongly assigned is highly improbable because of the identification of $RdTh(Th^{228})$ from electron-capture branching of $Pa^{228}$,

of ThX(Ra$^{224}$) from electron-capture branching of Ac$^{224}$ and ThC'(Po$^{212}$) from the β$^-$-decay of ThC(Bi$^{212}$). All in all, it appears to be highly unlikely that ThA(Po$^{216}$) is β$^-$-unstable and therefore the 7.57-Mev alpha-group in Tn must arise from some mechanism not involving At$^{216}$. The At$^{216}$ could not come from Fr$^{220}$ formed by β$^-$-branching of Em$^{220}$ since Em$^{220}$ is surely β$^-$-stable. Perhaps some such explanation as that offered by Feather[35] for the observation of Karlik and Bernert is correct.

These examples of the use of alpha-decay data may serve to indicate some of the methods which have found application while in the following are presented some ideas on beta-stability, the transuranium elements and the rare earth region.

Beta-Stability in the Heavy Elements.-- As illustrated by the discussion of whether or not At$^{216}$ could arise from the thorium radioactive family it is often important to be able to predict beta-stability. Many of the short-lived alpha-emitters are beta-unstable but this instability is not noted because of overwhelming competition by alpha-decay. Nevertheless, the degree of beta-instability can often be calculated by means of closed decay cycles and conversely the beta-energies can serve as important links in calculating alpha-energies and in predicting branched decay. No attempt will be made in this report to show the calculations of beta-decay energies and Table 1 tells only whether or not a nuclide is known to be beta-stable or predicted to be so. Predictions are in parentheses. A recent table of Biswas and Mukherjee[36] listing types of instability, differs in some instances from Table 1 and does not give predictions of unobserved modes of instability or for unknown species.

The greatest uncertainties in predictions of beta-stability involve choosing which isotopes, if any, of astatine and francium are beta-stable. Measurements in this laboratory[8] on the alpha-energy of At$^{216}$ gave 7.79 Mev which when used with the alpha-energy of Po$^{216}$(ThA) and the decay energy of Pb$^{212}$(ThB) allows one to calculate that At$^{216}$ is unstable with respect to electron capture to Po$^{216}$ by 0.4 Mev. In a similar fashion, making use of the newly measured[8] alpha-energy of At$^{214}$, it is

## Table 1

### Beta-Stability in the Heavy Elements*

| Element | Beta-Stable | EC Unstable | $\beta^-$-Unstable | EC and $\beta^-$-Unstable |
|---|---|---|---|---|
| bismuth | $Bi^{209}$ | $Bi^{197}-Bi^{206}$, $(Bi^{207})$ | $Bi^{210}-Bi^{214}$ | $(Bi^{208})$ |
| polonium | $Po^{208}$, $Po^{210}$, $Po^{211}$ $(Po^{212})$, $(Po^{213}?)$ $(Po^{214})$, $(Po^{216})$ | $Po^{202}$, $(Po^{203})$, $Po^{204}-$ $Po^{207}$, $(Po^{209})$ | $Po^{215}$, $(Po^{217})$ $Po^{218}$ | |
| astatine | | $At^{204}-At^{211}$, $(At^{213}?)$ | $(At^{215}?)$ $(At^{217}, At^{218})$ | $(At^{212}?)$ $(At^{214})$ $(At^{216})$ |
| emanation | $(Em^{212}?)$, $(Em^{214})$ $(Em^{215}?)$, $(Em^{216}-Em^{218})$, $Em^{220}$, $Em^{222}$ | $(Em^{213})$ | $(Em^{219}?)$, $(Em^{221})$ | |
| francium | $(Fr^{219}?)$ | $(Fr^{218})$ | $(Fr^{221})$, $Fr^{223}$ | $(Fr^{220}, Fr^{222})$ |
| radium | $(Ra^{218}, Ra^{220}, Ra^{221})$, $Ra^{222}-Ra^{224}$, $Ra^{226}$ | $(Ra^{219}?)$ | $Ra^{225}$, $Ra^{227}$, $Ra^{228}$ | |
| actinium | $Ac^{225}$ | $(Ac^{222})$, $Ac^{223}$ | $Ac^{226}$, $Ac^{227}$, $Ac^{228}$ | $(Ac^{224}, Ac^{226})$ |
| thorium | $(Th^{224}, Th^{226})$, $Th^{227}-Th^{230}, Th^{232}$ | $(Th^{223})$, $Th^{225}$ | $Th^{231}$, $Th^{233}$, $Th^{234}$ | |
| protactinium | $Pa^{231}$ | $(Pa^{226})$, $Pa^{227}-Pa^{229}$ | $Pa^{232}$, $Pa^{233}$, $Pa^{234}$ | $Pa^{230}$, $(Pa^{232})$ |
| uranium | $U^{230}$, $U^{232}-U^{235}$, $(U^{236})$, $U^{238}$ | $(U^{227})$, $U^{228}$, $U^{229}$, $U^{231}$ | $U^{237}$, $U^{239}$ | |
| neptunium | $Np^{237}$ | $Np^{231}$, $(Np^{232})$ $Np^{233}-Np^{235}$ | $Np^{238}$, $Np^{239}$ | $(Np^{236}, Np^{238})$ |
| plutonium | $Pu^{236}$, $Pu^{238}-Pu^{240}$, $(Pu^{242}, Pu^{244})$ | $(Pu^{232}, Pu^{233})$, $Pu^{234}$ $(Pu^{235})$, $Pu^{237}$ | $Pu^{241}$, $(Pu^{243})$ | |
| americium | $Am^{241}$, $(Am^{243}?)$ | $Am^{238}-Am^{240}$ | $Am^{242}$, $(Am^{244})$ | $(Am^{242})$ |
| curium | $Cm^{242}$, $(Cm^{244}-Cm^{246}$, $Cm^{248})$ | $Cm^{238}$, $(Cm^{239})$, $(Cm^{240}?)$, $Cm^{241}$, $(Cm^{243}?)$ | $(Cm^{247}, Cm^{249})$ | |
| element 97 | $(97^{247})$ | | | |

* Predictions are enclosed in parentheses.

possible to show that $Po^{214}$ (RaC') is beta-stable. Under such circumstances in which

both $Po^{216}$ and $Po^{214}$ are beta-stable it is usual that $Po^{213}$ would be expected to be

beta-stable, although there are a number of cases known in which there are two beta-

unstable even-odd isotopes of lower mass number than the heaviest beta-stable even-even

isotope. By closing a decay cycle involving $Po^{213}$, $At^{213}$, $Pb^{209}$, and $Bi^{209}$, it may be

shown that $At^{213}$ must have an alpha-decay energy in excess of 9.20 Mev in order that

$At^{213}$ be heavier than $Po^{213}$. All that can be said from extrapolation in Figure 1 is

that $At^{213}$ could have an alpha-decay energy this high. As a result the question as to

whether or not $At^{213}$ is beta-stable cannot be resolved at present.

The other most likely beta-stable isotope of astatine is $At^{215}$. Here it

can be shown that $Po^{215}$ is ~ 0.8 Mev heavier than $At^{215}$ but it is not at all certain

that $At^{215}$ is stable with respect to $Em^{215}$.

Regarding this question of whether $At^{215}$ is beta-stable with respect to

$Em^{215}$ it is possible to show that this is a borderline case insofar as the estimations

are valid just as was found for the isobars $At^{213}$-$Po^{213}$. The following decay cycle

may be drawn with the known values for the decay energies indicated. One may estimate

the alpha-decay energy of $Em^{215}$ from Figure 1 and from this close the cycle to determine
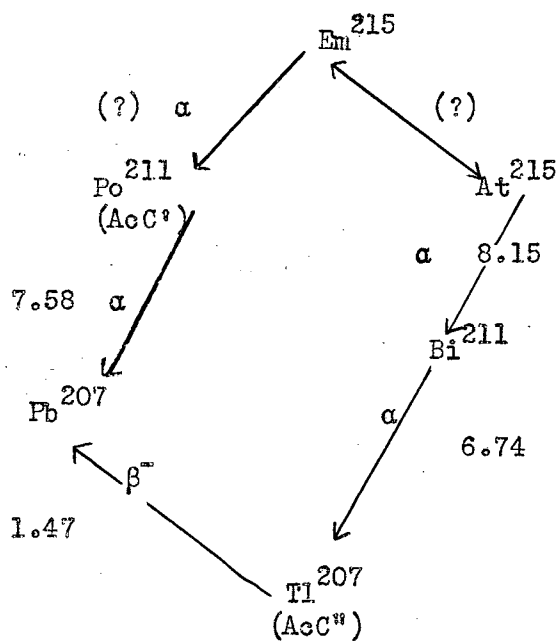
whether $Em^{215}$ or $At^{215}$ is heavier, or alternately, set these isobars equal and determine the alpha-energy which $Em^{215}$ would have to have to satisfy this condition. It is found accordingly that if the alpha-energy of $Em^{215}$ is less than 8.79 Mev then $Em^{215}$ is beta-stable and $At^{215}$ is beta-unstable. In examining Figure 1 it is seen that the requirement falls in the range of estimation (8.7 - 8.9) and it may be said that there is probably no more than 100 Kev difference between $Em^{215}$ and $At^{215}$. It should be mentioned that the most likely revisions that may be required in the measured values shown in the above decay sequences are in the direction of making $At^{215}$ heavier.

The possible beta-stable isotopes of astatine discussed above are $At^{213}$ and $At^{215}$. Higher isotopes of astatine such as $At^{217}$ are clearly beta-unstable.

Since there is no measurable isotope of astatine which can be proved to be beta-stable the question arises whether such exists or whether astatine is like two of the other missing elements, technetium (element 43) and prometheum (element 61), which probably have no beta-stable isotopes. In considering this point there are some remarkable analogies between these three elements which upon examination draw attention to a possible reason why no beta-stable isotopes exist. This has to do with the positions which the elements hold with respect to positions of stable configurations or "closed shells" in nuclear structure.

In the case of technetium it is necessary to examine the effect of neutron number 50 which apparently promotes added stability in nuclei in which it appears. It should be remembered that in general only one isotope of an odd element has a chance of being stable with respect to its even-element isobars. For technetium one might predict this isotope to be $Tc^{97}$ since it lies midway between $_{39}Cb^{93}$ and $_{45}Rh^{101}$. However, $Tc^{97}$ has two pairs of neutrons beyond the stable configuration of 50 neutrons and since such neutrons may be assumed to have abnormally low binding energies this may be just sufficient to cause it to be heavier than one of its isobars especially if the added assumption is made that for an even element the "closed neutron shell" has

a smaller effect on neutron binding energies beyond the shell. From this reasoning

all that can be said is that an odd element near the upper limit of elements which

have a stable isotope with the neutron number of abnormal stability should be affected

but not enough is known about the fine points of nuclear binding to predict whether

the affected element should be columbium (41) or technetium (43).

Prometheum (element 61) is in an analogous position with respect to neutron

number 82. Here there is a long list of stable nuclei, namely, $_{54}Xe^{136}$, $_{56}Ba^{138}$,

$_{57}La^{139}$, $_{58}Ce^{140}$, $_{59}Pr^{141}$, $_{60}Nd^{142}$, $_{62}Sm^{144}$. The remarkable stabilization of 82

neutrons allows both $La^{139}$ and $Pr^{141}$ to be beta-stable even though they differ by just

two protons and is presumably responsible for the absence of any stable isotope in the

next odd element, prometheum, through reasoning similar to that used for technetium.

The next apparent stabilizing neutron number occurs in the region of lead

and is presumably the number 126. In the heaviest elements about two neutrons are

added for each proton in building from element to element so it is not expected that

there should be as many elements each with a stable isotope with the same neutron number

as among the lighter elements. For neutron number 126, the known beta-stable nuclei

are $_{82}Pb^{208}$, $_{83}Bi^{209}$, and $_{84}Po^{210}$. The interesting possibility has already been

considered that $_{86}Em^{212}$ may be beta-stable. As is the case for neutron numbers 50 and

82 it may be expected that an odd element in this region will have no beta-stable

isotope, and it is postulated here that astatine (element 85) is that element.

The only possible beta-stable isotopes of francium are $Fr^{219}$ and $Fr^{221}$

since $Fr^{223}$ is known to decay to $Ra^{223}$ (AcX). It is not possible to say for sure

whether or not $Fr^{219}$ is beta-stable although tentatively it is decided to be so.

The question to be decided is which of the isobars $Fr^{219}$ and $Em^{219}$ (An) is heavier

since $Ra^{219}$ is almost surely unstable toward electron capture. A cycle containing

these isobars and including $At^{215}$, $AcA^{215}$, $AcB^{211}$, $AcC^{211}$ may be set up using values

shown in Figure 1 (see Table 3) for the alpha-energies and 1.40 for the disintegration

energy for $AcB^{211}$. When this is calculated it is found that $Fr^{219}$ is beta-stable with

respect to $Em^{219}$(An) by about 300 Kev. In view of the arguments as to the beta-instability of $Em^{219}$ it is almost certain that $Em^{221}$ is a $\beta^-$-emitter so that the consideration of the beta-stability of $Fr^{221}$ revolve around whether or not $Ra^{221}$ is electron capture unstable. Closing a cycle involving $Ra^{221}$, $Fr^{221}$, $Em^{217}$, $Po^{213}$, $At^{217}$ and $Bi^{213}$ it is found that $Fr^{221}$ is 100 Kev heavier than $Ra^{221}$, a difference which precludes a decision in view of the uncertainties in some of the data. Tentatively, the only beta-stable isotope of francium will be taken to be $Fr^{219}$.

Transuranium Elements.-- For the beta-stable nuclides and those with neutron excess the alpha-decay energies between comparable nuclei progressively increase above uranium. This effect has already been pointed out in the discussion of Figure 4. This means that comparable nuclei (in their position with regard to the center of beta-stability for each element) will show increased alpha-energy and shorter half-life in going above uranium. If one were to assume that alpha-decay is the only means by which very heavy nuclei could spontaneously degrade to lighter nuclei this effect can be deduced independently of measured alpha-energy values by the fact that transuranium elements have not survived through geological time.

To illustrate this trend the alpha-energies of analogous nuclei are listed in Table 2 for this region for the even elements. Nuclides in the same horizontal row are considered analogous. A few values obtained by interpolation and extrapolation are included and appear in parentheses.

Table 2

Trends of Alpha-Energies of "Comparable Nuclei".

| | | |
|---|---|---|
| $U^{232}$ - 5.40 | $Pu^{238}$ - 5.60 | $Cm^{244}$ - 5.88 |
| $U^{233}$ - 4.89 | $Pu^{239}$ - 5.24 | $Cm^{245}$ - (5.7) |
| $U^{234}$ - 4.84 | $Pu^{240}$ - 5.2 | $Cm^{246}$ - (5.6) |
| $U^{235}$ - 4.63 | $Pu^{239}$ - 5.24 | |
| | or | |
| | $Pu^{241}$ - 5.1 | |
| $U^{236}$ - (4.6) | $Pu^{242}$ - (4.9) | |

One of the uses to which these correlations may be put is the prediction of the nuclear properties of transcurium isotopes. The lightest beta-stable isotope of element 98 is $98^{246}$ or possibly $98^{248}$. By following the trends noted $98^{246}$ and $98^{248}$ might be predicted to have alpha-energies of 6.8 and 6.6 Mev, respectively. The correlations of half-lives of even-even isotopes as shown in Figure 6 allow one to extrapolate to element 98 and predict alpha half-lives of a few days and about one month, respectively, for the isotopes $98^{246}$ and $98^{248}$.

Alpha-Radioactivity in the Rare Earth Region.-- It is probably a safe assumption to make that alpha-active isotopes of almost all elements in the upper half of the periodic table may be produced by removal of a sufficient number of neutrons. A possible pattern which such alpha-emitters would follow has been suggested by Kohman.[19] In the region immediately below lead the departure from the region of beta-stability in which alpha-decay rate becomes appreciable is not so great but that the electron-capture half-lives allow one to observe the alpha-activity. The gold and mercury alpha-emitters would fall in this category and these are also analogous to the bismuth alpha-emitters around mass number 200. In lower regions of the periodic table where the general slope of the packing fraction curve is less favorable for alpha-emission one might expect to find appreciable alpha-decay rates only at a sufficient degree of instability toward electron capture or positron-decay that half-lives would be very short and detection of alpha-decay difficult. However, if there should be a localized region of steep slope in packing fraction then a moderate degree of neutron deficiency may enhance the effect sufficiently to produce readily observable alpha-emitters. Just such a region has probably been found recently in this laboratory.

Thompson et al.[23] have produced a number of alpha-active rare earths with half-lives in the range from a few minutes to a few days and alpha-particle energies in the range 4.2-3.1 Mev. Although these activities have not been positively assigned, it is likely that they belong to neutron deficient isotopes of gadolinium, terbium,

dysprosium, and holmium, but not samarium and lower rare earths. A most attractive hypothesis is that these nuclei owe their high potential toward alpha-emission to their position relative to the stable configuration of 82 neutrons just as the most energetic alpha-particles in the heavy element region come from nuclei decaying to the vicinity of 126 neutrons. The difference in the case of the rare earths is that these nuclides will in almost all cases lie outside of the limits of beta-stability on the neutron deficient side and the energies of alpha-emitters with 84 neutrons will be relatively lower than those with 128 neutrons because of the generally lower slope of the packing fraction curve. In view of these considerations, it would seem likely that the currently discovered group of alpha-emitters will include and cluster around such nuclides as $Gd^{148}$, $Tb^{149}$, $Dy^{150}$, and $Ho^{151}$. Perhaps alpha-emission will not be noted for isotopes more than two mass units above $Gd^{148}$ and about four mass units above $Ho^{151}$. One may turn to some previously available data for confirmatory evidence for this picture. It has long been noted that $Sm^{146}$, although surely beta-stable, is missing in natural samarium and presumed to be an alpha-emitter. This would indicate that samarium is the only element in this region for which an isotope with high alpha-energy falls within the limits of beta-stability. It would also be tempting to assign the long-lived alpha-emitter in natural samarium to $Sm^{147}$ or $Sm^{148}$ on this picture but the best direct evidence to date places the assignment at $Sm^{152}$ [37].

## ALPHA-DECAY RATE

On a simplified model of the alpha-decay process the alpha-particle is considered to exist as an entity within the nucleus of its decay product and its rate of emission is governed by the potential field of that nucleus. The factors which then determine the decay constant are the atomic number, the alpha-energy or velocity and the nuclear radius. For a particular decay energy, an increase in atomic number decreases the decay constant and decrease in nuclear radius decreases the decay constant through its effect in increasing the potential barrier. The quantum mechanical

treatment for the decay process resulted in the well-known Gamow formula[2] of which
there are a number of modifications and was remarkably successful in explaining the
great sensitivity of decay constant to decay energy. The effect of atomic number is
predicted to be much less striking and this is borne out by observation. The nuclear
radius cannot be independently evaluated with the accuracy required and the formula
has been used to calculate the "effective radius" which therefore has become a catch-
all for any quantitative shortcomings of the theory. As it turned out, the "effective
radius" undergoes rather large changes departing from the $A^{1/3}$ relationship by 25%
in some cases. One recognized factor in lengthening half-life which appears in the
effective radius is change in spin number in the decay process. However, it will be
seen that the present availability of much new data has shown up a number of regularities
including broad classes of nuclei with abnormally long half-lives and these cannot be
explained by spin changes. Rather, it is believed that the simplified one-body model
is inadequate in these cases and the process of creating the alpha-particles is slower
in certain nuclear types. These nuclear types include those which have an odd number
of neutrons, protons or both and in this sense alpha-emission is forbidden in nuclei
possessing non-zero spin beyond and independent of any effect due to spin change in the
decay process.

References to most of the data which have been used in these correlations
will be found in the compilation already cited.[3]    Table 3 shows the alpha-energies
and alpha-decay half-lives for all of the alpha-groups used. Those which are not
covered in reference 3 or for which additional data are available are discussed in
the Appendix. The data in Table 3 are entered on a series of diagrams (Figures 6, 7,
8, and 9) in which the curves drawn are taken to represent the energy vs. half-life
relations for non-forbidden alpha-decay processes. These curves, as will be explained,
are defined by the even-even nuclei. Other types are, as a rule, comparatively for-
bidden in their alpha-decay and the last column in Table 3 indicates the factor of departure
between the observed half-life and the predicted half-life if the decay process were

not forbidden.

The Even-Even Nuclei.-- In Figure 6 are plotted the data for the even-even

nuclei relating the half-life and energy in which lines of constant Z are shown.

With minor irregularities the points fall on a series of parallel lines. There is

still sufficient uncertainty in a few of the points which produce the irregularities

(e.g. $Pu^{240}$, $U^{228}$) to allow the possibility that the definition of the curves may

be even better when the measurements have been refined.

Of the known even-even nuclei only $Po^{210}$, $Po^{208}$, $Po^{206}$, and $Po^{204}$ fall

sharply out of line with others and for reasons which will appear below they have been

entered on another plot (Figure 9). It is probable that these species and/or their

decay products have abnormally low nuclear radii as compared with the heaviest polonium

isotopes and most isotopes of higher elements. These are the polonium isotopes which

in Figure 1 can be seen to lie in the region following the sharp break in the mass

number vs. energy curve and as would be expected the half-life vs. energy relation

also shows a discontinuity. However, even taking into account the decreased decay

energy these isotopes have abnormally long half-lives, another reflection of the shrink-

age in nuclear radius is this region. These will be discussed further below where

other isotopes with abnormally long half-life are considered.

Throughout the remainder of this paper these curves for the even-even nuclides

form a baseline for comparison of the half-life energy relationships of other nuclear

types. This family of curves, in form and without regard to nuclear type, falls

directly out of the quantitative treatment of the alpha-decay process in which the

parameter Z is held constant for each curve and nuclear radius is assumed to be a

smoothly varying function of mass number. This method for treating alpha-decay data

has already been applied by Berthelot[38] and Biswas[39] who attempted to fit all

nuclear types to the curves. In later discussions in this paper it will be seen that

rather good quantitative agreement may be brought about between these data for the

even-even nuclides and the Gamow formula by making reasonable assumptions for nuclear

radius. In the immediately following sections it will be seen that nuclear types other than even-even do not fall on this family of curves.

The position of the curve for polonium isotopes in Figure 6 is worthy of note since there is evidence from the calculations that $Po^{212}$ and possibly $Po^{214}$ are showing the effect of decrease in nuclear radius which lengthens the half-life and which becomes pronounced for $Po^{210}$ and lighter polonium isotopes. To this extent we have been inconsistent in placing the baseline curve for polonium through these points ($Po^{212}$ and $Po^{214}$).

An important part of the discussion in following sections on the nuclides with odd nucleons is the part which fine structure plays in the degree of prohibition of alpha-decay. There is an important distinction in this respect between the even-even nuclei and all others. There are three well defined cases of fine structure among the even-even nuclides and within the degree of reliability of the experimental data all groups fall on the curves of Figure 6. That is to say, the partial alpha half-lives for the two groups of each of the nuclides $Ra^{226}$, $Th^{230}$(Io) and $Th^{228}$(RdTh) are what would be expected for the respective energies of the groups. It will be seen that for nuclides with odd nucleons there is a pattern of prohibition of alpha-decay in which the ground state transition is the most highly forbidden.

Even-Odd Nuclei.-- When the points for nuclei of even Z and odd number of neutrons are entered on a half-life vs. energy plot it is found that almost all lie significantly above the curves for the respective elements as determined by the nuclei with even number of neutrons. These are shown in Figure 7 in which the even-odd nuclei are indicated by their symbols while the reference lines are curves for the even-even nuclei based on those in Figure 6.

Of especial interest are a number of cases in which the even-odd nuclei emit two or more alpha-groups. For these the partial alpha half-lives have been calculated (Table 3) and plotted with the appropriate energy values. It will be noted that in such cases one or more of the shorter range groups are less forbidden by the criterion

adopted than is the alpha-group of the ground state transition. A graphic illustration of this effect is seen in the three alpha-groups of $Th^{229}$ in which comparison with the curve of Figure 6 for the even-even thorium isotopes shows that the 5.14 Mev group (ground state transition) is 350-fold forbidden while the 5.04 Mev and 4.84 Mev groups are respectively 45-fold and 3-fold forbidden. All of the other cases of fine structure are qualitatively similar to that of $Th^{229}$ in the sense that the ground state transition is most highly forbidden and a low energy group may be relatively non-forbidden.

It is significant that all of the even-odd nuclei (that can be prepared in a state of purity which permits examination) may have important non-ground state transitions as evidenced either by direct observation of the alpha-groups or by the appearance of gamma-radiation in high abundance. The case of $Th^{229}$ in the 4n + 1 family, for which three alpha-groups have been measured, has already been mentioned and in this category of well defined multiple fine structure also lie $Th^{227}$ (RdAc), $Ra^{223}$ (AcX), and $Em^{219}$ (An) of the actinouranium family.

Because of the abnormally long partial alpha half-lives of the ground state transitions of the even-odd nuclei cited it is reasonable to suppose that there are other cases in which the ground state transitions have not been observed. There was good reason to suspect that $U^{235}$ was an example of this behavior as a gamma-ray of 160 Kev was found in high abundance and hence presumably in cascade with the only observed alpha-group (see Appendix for references and details). We have now found a group in 10 $\pm$ 1% abundance corresponding to the ground state transition which in unseparated or partially separated $U^{235}$ is obscured by the $U^{234}$ alpha-particle (see Appendix). A consequence of these observations is that while the 4.48 Mev transition is somewhat greater than 10-fold forbidden, the ground state transition (4.64 Mev) is 1000-fold forbidden. In view of these findings a re-evaluation of the specific activity and half-life of $U^{235}$ is in order especially since the half-life is involved in calculations of the age of the earth.

With the behavior of $U^{235}$ in mind it would be interesting to examine the
relation of the gamma-radiation to the alpha-particles of other even-odd nuclei of
which $Pu^{239}$ and $U^{233}$ may serve as examples. It may be that here too the observed
alpha-particles do not correspond to the ground state transitions and that the ground
state transitions are highly forbidden. Both $U^{233}$ and $Pu^{239}$ have associated gamma-
radiation in moderate abundance but it is not yet known whether such transitions are
in cascade with the observed alpha-particles. Some indirect evidence to lend further
support to the view that the observed alpha-particles do not represent ground state
transitions is obtained from Figures 1 and 7. From Figure 1 it may be seen that
$Pu^{239}$, $U^{233}$, and $Ra^{223}$ appear to have abnormally low alpha-decay energies, that is,
their decay energies as shown are little greater than their respective higher isotopes
$Pu^{240}$, $U^{234}$, and $Ra^{224}$. The inference might follow that to these energies should be
added some gamma-ray energies as has been done for $U^{235}$ and $Am^{241}$. From Figure 7 it
may be seen that with respect to half-life vs. energy relationship the observed $Pu^{239}$
alpha-particle is comparable to the short range (main) group of $U^{235}$ and $U^{233}$ is much
like the shortest range group of $Th^{229}$ while the known alpha-groups of $Ra^{223}$ are like
the shorter range groups of $Th^{227}$. From these relationships of Figure 7, one might
infer that the ground state transitions of $Pu^{239}$ and $U^{233}$ are more highly forbidden
as are the corresponding groups of their decay products. The significance of the
half-life vs. energy relationship in the case of fine structure in alpha-decay will be
discussed further below.

It will be noted in Figure 7 that $Po^{213}$ and $Po^{215}$ fall on the polonium
curve and hence would appear to be cases of unprohibited alpha-decay. Before accepting
these as exceptions one must consider that the baseline in this region of the polonium
curve is defined by $Po^{212}$ and $Po^{214}$ and that these nuclei are themselves prohibited due
to shrinkage in nuclear radius (see section on <u>Quantitative Treatment of Alpha Decay</u>).
There is also the possibility that the observed alpha-particles of $Po^{213}$ and $Po^{215}$

do not represent ground state transitions.

Odd-Even and Odd-Odd Nuclei.-- The half-lives for these types are plotted in Figure 8 in which the same reference lines for the even-even nuclei are shown as in Figures 6 and 7. Interpolated between these curves are broken lines to represent the positions which the isotopes of odd Z elements would have if their alpha-decay rates, like the even-even nuclei, were not forbidden. It is seen that almost invariably these types are forbidden and that the odd-odd species show greater departures from their curves than do the odd-even ones. The quantitative bases for these generalizations may be found in Table 3.

For the odd-even species there is reason to doubt that the departures from the reference curves as shown in Table 3 and Figure 8 give a true picture of the forbidden character of the ground state transitions since, as has already appeared for the even-odd species, it is not certain that the observed alpha-particles represent these transitions. For example, the measured alpha-particle of 5.48 Mev for $Am^{241}$ almost surely does not correspond to the total decay energy since there is found a gamma-ray of 62 Kev in high abundance. If, as seems likely, there should be an unobserved alpha-group of 5.54 Mev, then it must have a very long partial half-life and its emission is highly forbidden. How many others of the odd-even type exhibit the same behavior as $Am^{241}$ is not known.

There are two recognized cases of fine structure in alpha-decay among the odd-even nuclei. These are $Pa^{231}$ and $Fr^{221}$. In both instances the lower energy group is the less highly forbidden, in fact, the 6.05 Mev group for $Fr^{221}$ does not appear to be prohibited.

In examining the data for the odd-even nuclei in Table 3 it may be noted that a large fraction of the partial alpha half-lives are calculated from very rare alpha-branching of predominant electron capture decay processes. There is considerable uncertainty in estimating the number of electron-capture events and the experience in this laboratory has indicated that it is most likely that they are underestimated.

This means that the true partial alpha half-lives are probably longer than has been tentatively accepted.

The single example of an odd-even nucleus for which both energy and alpha half-life should be reliable which is not forbidden is $At^{217}$. There is no apparent explanation for this exception to the general rule unless $At^{217}$ is another case in which the ground state transition is not measured.

It is apparent from the few examples of odd-odd species that alpha-emission in this type is forbidden. With the reservations necessary because of uncertainties in data as already discussed it is probable that odd-odd nuclei are more highly forbidden in their alpha-decay than are the even-odd and odd-even types.

Special Forbidden Species.-- The special significance of this group plotted in Figure 9 will be discussed below. Included are all known bismuth alpha-emitters and isotopes of francium, emanation, astatine, and polonium with 126 or fewer neutrons. In the case of polonium this group includes several isotopes of the even-even type and as already discussed should probably include $Po^{212}$ and $Po^{214}$.

Discussion of Forbidden Alpha-Decay.-- From the data shown in Figures 6, 7 and 8 it seems to be a general condition in alpha-radioactivity that nuclei with an odd neutron, proton or both show forbidden decay as compared with the even-even nuclei. This is particularly true of the ground state transitions in those cases in which more than one alpha-group has been measured. Where several alpha-groups are measured, the ground state transition is most highly forbidden and one or more of the shorter range groups is much less forbidden. In contrast, the few cases of fine structure in even-even nuclides indicate no prohibition for either group so that the partial alpha half-lives correspond to the respective alpha-energies.

According to the Gamow theory a factor which prohibits alpha-decay is spin change in the transition and of course any discontinuity in nuclear radius in the direction of abnormally low nuclear radius will appear as prohibited alpha-decay if the condition is not recognized. There is reason to believe that $Em^{212}$, $Fr^{212}$, $At^{211}$,

polonium isotopes of mass 214 and less, and perhaps all of the bismuth isotopes show the effect of shrinkage of nuclear radius. However, it is unreasonable also to attribute an important effect of nuclear radius to all of the heavier nuclides with odd protons and neutrons since, if anything, these species should exhibit somewhat greater radii than neighboring even-even isotopes. It is also not possible to explain the general abnormal half-lives of these types by means of spin changes.

According to the Gamow formulation the spin dependence is such that in order to explain alpha-decay abnormally long by a factor of 10 it would be necessary to invoke a spin change of about 5 units. While such large spin changes have been observed in beta transitions in which an odd-odd nucleus decays to an even-even type it would be unreasonable to expect such large changes, particularly as a general condition, in the case of alpha-decay in which parent and daughter nuclei are of the same type. In addition, considering a decay series such as the $(4n + 1)$ or $(4n + 3)$ series in which virtually all members show abnormal decay constants it would be necessary to postulate alternation of large and small spin numbers differing by several units each in proceeding down the decay chains. In one case of alpha-decay, namely that of $U^{235}$, the spin numbers of the ground states have been measured. For $U^{235}$ a value of 5/2 (or 7/2) was obtained[40] while for $Pa^{231}$ a value of 3/2 was measured[41], thus indicating a spin change of 1 or 2 units in the transition. In order to account for prohibition of the ground state transition of $U^{235}$ by spin change alone it would be necessary that there be a change of about 10 units.

Since present alpha-decay theory does not explicitly account for the forbidden transitions in all of the broad categories in which they are observed a qualitative modification is suggested which is not out of harmony with the Gamow and Condon-Gurney theory. An examination of the Gamow formula shows it to consist of two parts; an exponential term describing the barrier penetration and a coefficient before the exponential term which is a slowly varying function and has been taken to be a constant.

This may be written in the form:

$$\lambda = Ce^{-f(v, Z, r)}$$

Here $\lambda$ is the decay constant, C is the coefficient mentioned and v, Z and r are the alpha-particle velocity, the atomic number and the effective nuclear radius. The constant C may be thought of as related to the frequency with which an alpha-particle in the parent nucleus encounters the potential barrier and numerically approximates the reciprocal of the length of time it takes the alpha-particle to cross nuclear dimensions. This model makes the tacit assumption that the alpha-particle which is emitted has been formed in the nucleus and that there is no prohibition toward assembling its components in any one nucleus as compared with any other. It is here that we suggest that the effect of an odd nucleon makes itself felt in slowing the assembly of an alpha-particle. The odd nucleon, presumably the one in the highest quantum state, must be a component of the emitted alpha-particle if the alpha-particle is to leave the nucleus with full kinetic energy; as a result it must pair with a lower lying nucleon of anti-parallel spin and perhaps in addition one or more of the remaining nucleons may have to change quantum states. If we assume that these processes require appreciable time we have the basis for forbidden decay in nuclei with odd nucleons. By the same token two odd nucleons should in general prohibit alpha-decay more than single odd nucleons and this would be borne out by comparison of the odd-odd nuclei with the odd-even or even-odd types in Table 3 and Figures 7 and 8. It would also follow from these ideas that in all of the forbidden classes lower energy alpha-groups might compete with or supplant the ground state transition since the lower lying nucleons are more likely to be paired and in any case should not be so prohibited in their assembly into an alpha-particle. As is observed, these groups should be less forbidden for their energies than the ground state transition.

The abnormality in the half-life vs. energy relationships for $Po^{210}$ and all of the naturally occurring bismuth isotopes as compared with other alpha-emitters has been pointed out a number of times and the effect has been attributed to a sudden shrinkage of nuclear radius in these species. It is worthwhile to attempt to correlate such abnormalities in nuclear dimensions as evidenced by alpha-decay properties with the new ideas on stable configurations in nuclear structure. For unambiguous interpretation in a region of rapidly changing nuclear radius bridged by an alpha-decay event it would be necessary to decide whether the increased barrier from a shrunken nucleus may be asribed to the parent or daughter nucleus. In the one-body model the alpha-particle is considered to be a body moving within the decay product nucleus and the barrier to which it is subjected is that of the decay product. This would mean, as an example, that the alpha-decay of bismuth isotopes is forbidden because the thallium decay products have abnormally low nuclear radii. It is probable that this model is not adequate and that the nucleus responsible for the potential barrier is a hybrid between parent and daughter nuclei.

As already mentioned all of the naturally occurring isotopes of bismuth show very highly forbidden alpha-decay ranging from several hundred-fold to several thousand-fold above the baseline curve. Part of this may now be attributed to odd nucleons in bismuth isotopes, but there is still a considerable degree of prohibition presumably due to nuclear radius effects. If we take a representative departure from the baseline curve as a factor of 500 and attribute 10 or 20 of this to odd nucleons, there is left a factor of 50 or 25 for the effect of decrease in nuclear radius. According to the Gamow formula a shrinkage of about 10% could account for a fifty-fold increase in half-life. In the following section are discussed more fully some of the quantitative aspects of alpha-decay theory and here it will be seen that there is good agreement with the theory for even-even nuclei except the lighter even-even isotopes of polonium, $Po^{212}$, $Po^{210}$, and $Po^{208}$. In these cases we attribute the entire prohibition of alpha-emission to nuclear radius effects since other even-even nuclides

removed from the region of 82 protons and 126 neutrons show rather good agreement. For $Po^{210}$ and $Po^{208}$, the degree of prohibition is such that a postulated shrinkage of almost 10% is necessary to account for it. Since the bismuth isotopes when divested of the effects of odd nucleons show similar prohibition it is only necessary to postulate nuclear shrinkage of the same order, namely, 10% or less.

With regard to the new highly neutron deficient bismuth alpha-emitters it is not possible to say whether there is any trend away from that noted for the heavy bismuth alpha-emitters because the branching ratios between alpha-emission and electron capture are not known accurately and the calculated alpha half-lives shown in Table 3 must be considered only as rough approximations. The two-minute period assigned to $Bi^{197}$ is only about 10-fold forbidden if it decays mainly by alpha-emission. There is no evidence on this point, however. On the other hand the best measured alpha-branching of 25-min. $Bi^{199}$ shows its alpha-decay to be highly forbidden. In attributing and explaining these effects for bismuth in terms of stability of closed shells in nuclear structure perhaps the best that can be done at present is to say that there is abnormally strong binding energies of one or more protons before the closed shell 82 and that for neutrons there are high binding energies for a sizeable number before 126.

With the marked retardation of alpha-decay for bismuth isotopes one might expect that lead isotopes (Z = 82) would accentuate this trend. As a matter of fact no lead alpha-activity has been observed even for highly neutron deficient species in corresponding position to the light bismuth isotopes in which alpha-activity does again manifest itself. This might receive adequate explanation by assuming low decay energy for lead isotopes but in addition there could be extremely long half-lives for the decay energies. It is perhaps significant of the position of lead that Thompson et al[23] have observed alpha-activity again when this region

has been crossed, that is, for highly neutron deficient species of gold (Z = 79)

and mercury (Z = 80).

Besides the bismuth isotopes there is another class that seems to be for-

bidden in alpha-decay for reasons of abnormal nuclear radius. These are the polonium

isotopes of mass 210 and lower, $At^{211}$ and $Em^{212}$. The significant point here is

that all of these have 126 neutrons or less. It will be noted that these are

the nuclei in Figure 1 that show a sharp departure in the mass number vs energy

regularities of the heavier isotopes. In this region even the even-even nuclei

appear to be forbidden in their alpha-decay.

In Figure 3 an attempt has been made to demarcate the regions of abnormal

nuclear stability. For this purpose use was made of two observations, namely,

that all bismuth isotopes show a high degree of prohibition in alpha-decay and

that all nuclides with 126 or less neutrons also show this property. The implica-

tion of the abnormally long half-lives in both cases is that there are discontinui-

ties in nuclear radius. With regard to nuclei having 126 neutrons or less, it

seems fairly clear that there is a region below 126 neutrons in which nuclear

binding varies only slightly but that as this neutron number is exceeded there is

a sharp decrease in binding energy. This effect shows up as a maximum alpha-energy

for a nucleus with 126 neutrons which decays to one with 126 neutrons as is the

case for $Bi^{211}$, $Po^{212}$ and presumably $At^{213}$ and higher nuclei of the type. Likewise

nuclei with 126 neutrons and less have low alpha-energy, and what is of equal

importance, the half-lives are abnormally long for the alpha-energies. If several

neutrons beyond 126 are bound relatively weakly then the alpha-decay energies of

nuclei with 127 neutrons should be between those with 126 and 128 neutrons because

the alpha-particle here carries off one neutron which is firmly bound and one

which is loosely bound. This would account for the observed alpha-energy of $Po^{211}$

(AcC') and leads to the prediction of that for $At^{212}$ as shown in Figure 1. Another

effect should be observed for these nuclides, namely, the highly forbidden nature

of alpha-decay so prominent for nuclei with 126 and less nuclei should begin to

show up. For $Po^{211}$ this would lead to the prediction that the half-life might be

as long as 50 milliseconds, some ten-fold greater than that predicted presumably

from the Geiger-Nuttall relations.[42]

This discussion of the effect of 126 neutrons on the energy surface

probably applies equally well to the proton number 82 but in this case the almost

total absence of measurable alpha-decay potential immediately below bismuth makes

any speculation of dubious value. However, the discussion of the effect on

alpha-decay of one nucleon beyond a closed shell as applied above with scanty data

to the neutron number 127 has more abundant examples in the proton number 83.

All of the alpha-energies of bismuth isotopes are sharply lower than those of

polonium as compared with the displacement between succeeding elements and the

decay rates show a degree of prohibition beyond that expected for nuclides with

odd nucleons.

Quantitative Treatment of Alpha Decay.-- From preceding discussions it

seems obvious that if there is to be quantitative agreement between experimental

data and existing theory that this agreement can only come for the even-even nuclides.

Preliminary calculations* with these indicate that agreement is indeed good over

a wide range of mass number and atomic number. Thus the shape and spacing of the

curves shown in Figure 6 are reproduced rather faithfully by the single body

theory of alpha-emission as has already been indicated by Biswas[39] and others

who have in addition attempted to fit all nuclear types on the same curves.

Since the nuclear radius is the only parameter in the formula which is

not known it is tempting to use curve fitting for the even-even nuclides as a

means of determining a function which will describe the nuclear radius in the

heavy element region. No comprehensive computations have yet been made, but preliminary

attempts would indicate that the simple function $r = 1.48\ A^{1/3} \cdot 10^{-13}$ cm, will fit

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the data better than a function such as this with an additive term to express the
radius of the alpha-particle or the range of its forces as has been suggested.[43,44]
One may calculate conversely the explicit value for the nuclear radius for each
even-even nuclide which is necessary to give the observed value for the decay
constant and to determine the deviation of each such radius from the rule, $r =
1.48 \ A^{1/3} \cdot 10^{-13}$ cm. In about a dozen such calculations for nuclides from emanation
to curium, the mean deviation encountered was about 1% which is considered good
agreement when it is considered that often uncertainties in alpha-energy are
reflected by greater differences in nuclear radius. Refinement of energy measure-
ments and correlation of more extensive data will show whether there are trends
away from this simple formulation for nuclear radius.

Calculations of nuclear radius have also been made for six of the even-
even isotopes of polonium for which reliable decay energies and half-lives are
available. It has long been apparent in the decay of $Po^{210}$ and possible all
bismuth isotopes that a shrinkage in nuclear radius is at play in lengthening
the half-lives. In the case of the bismuth isotopes we must attribute part of
the prohibition to the general effect of odd nucleons, part possibly to spin
changes and part to nuclear radius effects. However, for the even-even polonium
isotopes any appreciable prohibition should be ascribed to the abnormality of
nuclear radius. In the single body theory of alpha-decay it is assumed that the
nuclear radius in effect is that of the alpha-decay product. In the case of polonium
alpha-emitters one would then be "measuring" the nuclear radii of the corresponding
lead isotopes. Whether or not there is any meaning in ascribing the parameter
of distance in alpha-decay explicitly to the radius of the product nucleus, it
is almost certain that the binding energy and hence radius of the product nucleus
is an important factor in the energy vs half-life relationship. For the decay
of polonium isotopes, one may attribute prohibition of alpha-decay to shrunken
nuclear radii of the lead daughter isotopes associated with the stable configura-
tion of 82 protons. For $Po^{212}$ and lighter isotopes, the decay products have 126

or less neutrons, and this should increase further the prohibition because of the further decrease in effective nuclear radius. Finally, if we assume that the radius of the parent nucleus is likewise a determinative factor in the effective radius for alpha-emission, polonium isotopes of mass number 210 and lower should have still more highly forbidden alpha-decay since the parent nuclei also have 126 neutrons or less. It is interesting to note that the calculated radii for the even-even polonium isotopes deviate in the manner expected from radii determined by the simple $A^{1/3}$ function. While the $Po^{218}$ radius is found to be low by only 1.4% the isotopes $Po^{216}$, $Po^{214}$, $Po^{212}$, $Po^{210}$ and $Po^{208}$ show deviations respectively of 2.1%, 2.8%, 5.5%, 8.1% and 9.0%.

It may be mentioned that further dissociation of factors which effect the nuclear radius may be obtained from observations on nuclides such as $Em^{212}$. Here both parent and daughter nuclei have 126 or less neutrons but the daughters have 84 protons rather than the 82 for the daughters of polonium isotopes. It will be noted that, perhaps fortuitiously, $Em^{212}$ is less forbidden than is $Po^{210}$ and $Po^{208}$.

## Table 3

### Alpha Energies and Half Lives

| Nucleus | Alpha Energy | | Abundance of α Group | Measured Half-Life | Alpha Branching Ratio | Alpha Half-Life | Departure* Factor |
|---|---|---|---|---|---|---|---|
| | | | | Even-even species | | | |
| $Cm^{242}$ | 6.18 | | | 150 days | | 150 days | |
| $Cm^{240}$ | 6.37 | | | 26.8 days | >0.8 | ~30 days | |
| $Pu^{240}$ | 5.2 | | | ~6000 yrs | | ~6000 yrs | |
| $Pu^{238}$ | 5.60 | | | 92 yrs | | 92 yrs | |
| $Pu^{236}$ | 5.85 | | | 2.7 yrs | | 2.7 yrs | |
| $Pu^{234}$ | 6.26 | | | 8.5 hr | ~0.03 | ~10 days | |
| $U^{238}$ | 4.25 | | | $4.51 \times 10^9$ yrs | | $4.51 \times 10^9$ yrs | |
| $U^{234}$ | 4.84 | | | $2.35 \times 10^5$ yrs | | $2.35 \times 10^5$ yrs | |
| $U^{232}$ | 5.40 | | | 70 yrs | | 70 yrs | |
| $U^{230}$ | 5.96 | | | 20.8 days | | 20.8 days | |
| $U^{228}$ | 6.83 | | | 9.3 min | 0.8 | 12 min | |
| $Th^{232}$ | 4.05 | | | $1.39 \times 10^{10}$ yrs | | $1.39 \times 10^{10}$ yrs | |
| $Th^{230}$ | 0 4.68 | I 4.61 | 0.80 0.20 | $8.0 \times 10^4$ yrs | | $1.0 \times 10^5$ $4.0 \times 10^5$ | |
| $Th^{228}$ | 0 5.52 | I 5.43 | 0.70 0.28 | 1.90 yrs | | 2.64 6.79 | |
| $Th^{226}$ | 6.41 | | | 30.9 min | | 30.9 min | |
| $Ra^{226}$ | 0 4.88 | I 4.68 | .91 .09 | 1622 yrs | | 1780 $1.8 \times 10^4$ | |
| $Ra^{224}$ | 5.78 | | | 3.64 days | | 3.64 days | |
| $Ra^{222}$ | 6.62 | | | 38 sec | | 38 sec | |
| $Em^{222}$ | 5.59 | | | 3.83 days | | 3.83 days | |
| $Em^{220}$ | 6.39 | | | 54.5 sec | | 54.5 sec | |
| $Em^{218}$ | 7.25 | | | 0.019 sec | | 0.019 sec | |
| $Po^{218}$ | 6.12 | | | 3.05 min | ~1 | 3.05 min | |
| $Po^{216}$ | 6.89 | | | 0.158 sec | | 0.158 sec | |

* Factor by which measured half-life is greater than value for same Z and $E_\alpha$ taken off curves defined by even-even nuclides.

Table 3 (cont.)

| Nucleus | Alpha Energy | Abundance of α Group | Measured Half-Life | Alpha Branching Ratio | Alpha Half-Life | Departure* Factor |
|---|---|---|---|---|---|---|
| $Po^{214}$ | 7.83 | | $1.5 \times 10^{-4}$ sec | | $1.5 \times 10^{-4}$ sec | |
| $Po^{212}$ | 8.95 | | $3.0 \times 10^{-7}$ sec | | $3.0 \times 10^{-7}$ sec | |

Even-odd species

| Nucleus | Alpha Energy | Abundance of α Group | Measured Half-Life | Alpha Branching Ratio | Alpha Half-Life | Departure* Factor |
|---|---|---|---|---|---|---|
| $Pu^{241}$ | 5.1(calc.) | | ∼10 yrs | $2 \times 10^{-5}$ | $5 \times 10^{5}$ yr | 9 |
| $Pu^{239}$ | 5.24 | | | | $2.4 \times 10^{4}$ yr | 4 |
| $U^{235}$ | 4.64 | 0.10 | $7.07 \times 10^{8}$ yr | | $7.1 \times 10^{9}$ yr | 1000 |
| | 4.48 | 0.90 | | | $7.7 \times 10^{8}$ yr | 13 |
| $U^{233}$ | 4.90 | | $1.6 \times 10^{5}$ yr | | $1.6 \times 10^{5}$ yr | 2 |
| $U^{231}$ | 5.6(est) | | 4.2 days | $6 \times 10^{-5}$ | 200 yr | 40 |
| $U^{229}$ | 6.53 | | 58 min | 0.2 | 5 hr | 9 |
| $Th^{229}$ | O 5.14 | 0.1 | 7000 yr | | $7 \times 10^{4}$ yr | 350 |
| | I 5.04 | 0.2 | | | $3.5 \times 10^{4}$ yr | 45 |
| | II 4.94 | 0.7 | | | $10^{4}$ yr | 3 |
| $Th^{227}$ | O 6.16 | 0.20 | 18.6 d | | 93 days | 300 |
| | I 6.13 | 0.038 | | | | |
| | II 6.10 | 0.25 | | | 74.5 days | 100 |
| | III 6.08 | 0.038 | | | | |
| | IV 6.03 | 0.013 | | | | |
| | V 5.97 | 0.025 | | | | |
| | VI 5.92 | 0.013 | | | | |
| | VII 5.87 | 0.20 | | | 93 days | 10 |
| | VIII 5.84 | 0.038 | | | | |
| | IX 5.82 | 0.15 | | | 124 days | 8 |
| | X 5.77 | 0.025 | | | 745 days | 25 |
| $Th^{225}$ | 6.68 | | 7.8 min | 0.9 | 8.7 min | 5 |
| $Ra^{223}$ | O 5.82 | 0.55 | 11.2 days | | 20.2 days | 10 |
| | I 5.71 | 0.36 | | | 31 days | 4 |
| | II 5.63 | 0.90 | | | 124 days | 5 |
| $Ra^{221}$ | 6.83 | | 31 sec. | | 31 sec. | 7 |
| $Em^{219}$ | O 6.94 | 0.83 | 3.92 sec | | 4.7 sec | 14 |
| | I 6.68 | 0.085 | | | 46 sec | 10 |
| | II 6.56 | 0.085 | | | 46 sec | 3 |
| $Em^{217}$ | 7.88 | | $10^{-3}$ sec | | $10^{-3}$ sec | 4(?) |

Table 3 (cont.)

| Nucleus | Alpha Energy | Abundance of α Group | Measured Half-Life | Alpha Branching Ratio | Alpha Half-Life | Departure* Factor |
|---|---|---|---|---|---|---|
| $Po^{215}$ | 7.50 | | $1.83 \times 10^{-3}$ sec | $\sim 1(?)$ | $1.8 \times 10^{-3}$ sec | 1 |
| $Po^{213}$ | 8.49 | | $4 \times 10^{-6}$ sec | | $4 \times 10^{-6}$ sec | 1 |
| | | | Odd-even species | | | |
| $Am^{241}$ | I 5.58 | | 490 yr | | 490 yr | 2 |
| $Am^{239}$ | 5.87 | | 12 hr | $10^{-4}$ | 13.7 yr | 3(?) |
| $Np^{237}$ | 4.85 | | $2.2 \times 10^{6}$ yr | | $2.2 \times 10^{6}$ yr | 4 |
| $Np^{235}$ | 5.15 | | 435 days | $5 \times 10^{-5}$ | $2.5 \times 10^{4}$ yr | 4(?) |
| $Np^{233}$ | 5.65 | | 35 min | $10^{-5}$ | 10 yr | 1.0(?) |
| $Pa^{231}$ | O 5.10 | 0.87 | $3.43 \times 10^{4}$ yr | | $3.95 \times 10^{4}$ yr | 40 |
| | I 4.82 | 0.13 | | | $2.64 \times 10^{5}$ yr | 5 |
| $Pa^{229}$ | 5.79 | | 1.5 days | 0.01 | 150 days | 3(?) |
| $Pa^{227}$ | 6.57 | | 38 min | 0.8 | 50 min | 6 |
| $Ac^{227}$ | 5.04 | | 21.7 yr | 0.012 | 1810 yr | 7 |
| $Ac^{225}$ | 5.90 | | | | 10.0 days | 5 |
| $Ac^{223}$ | 6.76 | | 2.2 min | $\sim 1(?)$ | 2.2 min | 7 |
| $Fr^{221}$ | 6.45 | | 4.8 min | $\sim 1(?)$ | 4.8 min | 5 |
| $Fr^{219}$ | 7.44 | | 0.02 sec | | 0.02 sec | 3(?) |
| $At^{217}$ | 7.15 | | 0.021 sec | $\sim 1(?)$ | 0.021 sec | 1 |
| $At^{215}$ | 8.15 | | $10^{-4}$ sec | $\sim 1(?)$ | $10^{-4}$ sec | 4(?) |
| | | | Odd-odd Species | | | |
| $Am^{242}$ | 5.4(calc) | | | | $\sim 2 \times 10^{5}$ yrs | 20(?) |
| $Pa^{230}$ | 5.5(est) | | 17.7 days | $3.5 \times 10^{-5}$ | 1400 yrs | 250(?) |
| $Pa^{228}$ | 6.20 | | 22 hrs | $\sim 0.02$ | $\sim 50$ days | 150(?) |
| $Pa^{226}$ | 6.93 | | 1.7 min | $\sim 1(?)$ | 1.7 min | 6 |
| $Ac^{224}$ | 6.28 | | 2.9 hrs | 0.1 | 29 hrs | 40 |

Table 3 (cont.)

| Nucleus | Alpha Energy | | Abundance of α Group | Measured Half-Life | Alpha Branching Ratio | Alpha Half-Life | Departure* Factor |
|---|---|---|---|---|---|---|---|
| $Fr^{220}$ | | 6.81 | | 27.5 sec | $\sim 1(?)$ | 27.5 sec | 13 |
| $At^{216}$ | | 7.94 | | $10^{-3}$ sec | $\sim 1(?)$ | $10^{-3}$ sec | 10 |

Specially forbidden species
(Bismuth isotopes and others with 126 neutrons or less)

| Nucleus | Alpha Energy | | Abundance of α Group | Measured Half-Life | Alpha Branching Ratio | Alpha Half-Life | Departure* Factor |
|---|---|---|---|---|---|---|---|
| $Bi^{214}$ | 0 | 5.61 | 0.45 | 19.7 min | $4 \times 10^{-4}$ | 76 days | 400 |
| | I | 5.55 | 0.55 | | | 63 days | 150 |
| $Bi^{213}$ | | 5.97 | | 47 min | 0.02 | 39 hr | 350 |
| $Bi^{212}$ | 0 | 6.20 | 0.27 | 60.5 min | 0.337 | 11 hr | 1000 |
| | I | 6.16 | 0.70 | | | 4.3 hr | 600 |
| | IV | 5.70 | 0.011 | | | 272 hr (11.3d) | 150 |
| $Bi^{211}$ | 0 | 6.74 | 0.84 | 2.16 min | 0.997 | 2.6 min | 700 |
| | I | 6.38 | 0.16 | | | 13.5 min | 150 |
| $Bi^{210}$ | | 4.86 | | 5.0 days | $\sim 3 \times 10^{-7}$ | $5 \times 10^4$ yr | $10^4(?)$ |
| $Bi^{199}$ | | 5.58 | | 25 min | $\sim 2 \times 10^{-5}$ | 2 yr | 3000(?) |
| $Bi^{197}$ | | 6.3 | | 2 min | ? | >2 min | >10(?) |
| $Em^{212}$ | | 6.30 | | 23 min | ? | 23 min | 10 |
| $Fr^{212}$ | | 6.37 | | 19 min | .5 | 38 min | 16(?) |
| $At^{211}$ | | 6.00 | | 7.5 hr | 0.40 | 19 hr | 50 |
| $Po^{210}$ | | 5.40 | | 138 days | | 138 days | 30 |
| $Po^{209}$ | | 5.02 | | 200 yr | >0.9 | 200 yrs | 150 |
| $Po^{208}$ | | 5.24 | | 3 yr | | 3 yr | 30 |
| $Po^{207}$ | | 5.2 | | 5.7 hr | $\sim 10^{-4}$ | 7 yr | 50(?) |
| $Po^{206}$ | | 5.3 | | 9 days | 0.1 | 90 days | 6(?) |
| $Po^{204}$ | | 5.45 | | 4 hr | $\sim 10^{-4}$ | 5 yrs | 1000 |
| $Au^{<190}$ | | 5.3 | | 4.3 min | $10^{-4}$ | 1 mo. | 400(?) |

APPENDIX

Notes and Comments on Individual Activities

Most of the data used in this paper may be found in the compilation "Table of Isotopes"[3]. However there are a number of new isotopes not covered there, some of the data have been revised or amplified and some require further explanation in order to indicate the reliability for the purpose used here.

$Cm^{238}$ As listed[3] this is an alpha-emitter of 6.50 Mev with 2.5-hr. half-life. It is also surely unstable with respect to electron capture (see Table 1) and although the branching has not been determined it is probable that the measured half-life is essentially that of the electron capture process. As a result this isotope was not included in Figure 6 as part of the correlations of the even-even species.

$Cm^{240}$ This isotope of curium is also predicted to be unstable toward electron capture. However, the branching for this mode of decay has been shown[45] to be less than 20% so that little error is introduced in plotting the measured half-life as the alpha-decay half-life.

$Cm^{244}$ This is a long-lived alpha-emitter of 5.78 Mev alpha-energy discovered by S. G. Thompson[46] from the irradiation of $Am^{241}$ with 38 Mev helium ions. Thompson suggests alternate mass number assignments, $Cm^{243}$ and $Cm^{244}$, and the assignment to $Cm^{244}$ is favored here from some detailed considerations in the assignment of several curium isotopes.[45] From yield considerations involving considerable uncertainty, Thompson estimates the half-life as 10 years. This point has not been used in Figure 6 although it fortuitously falls about where expected.

$Am^{242}$ There is considerable uncertainty in the indicated half-life of the ground state of $Am^{242}$. The alpha half-life indicated in Table 3 is based on the $Np^{238}$ yield from alpha-decay assuming that $Am^{242}$ was formed in the same yield as the 16-hr. excited state, $Am^{242m}$, from the neutron capture by $Am^{241}$. There could be a substantial error from this source in either direction. The alpha-energy was determined by closing the cycle consisting of the beta-decay

energies of $Am^{242}$ and $Np^{238}$ and the alpha energy of $Cm^{242}$. The beta-decay energy of $Am^{242}$ used in closing the cycle is taken to be that of the beta-particle energy (0.6 Mev). If any of the gamma-radiation is in cascade with the beta particle the decay energy would be greater and this would increase correspondingly the $Am^{242}$ alpha-energy.

$Am^{241}$   The high abundance of soft gamma-radiation and L-x-rays led to the belief that the main alpha-group is in cascade with a gamma-ray. Preliminary alpha-gamma coincidence measurements[47] tend to confirm this assumption. The gamma-energy is therefore added to the energy of the alpha particle and recoil nucleus in arriving at the decay energy (5.64 Mev) shown in Figure 1.

$Am^{239}$   The listed[3] alpha-branching of ~0.1% has been redetermined[48] and a better value 0.01% has been used here in determining the alpha half-life.

$Pu^{241}$   The alpha half-life indicated in Table 3 is based upon the $U^{237}$ yield from alpha-decay estimating the amount of $Pu^{241}$ from yield considerations. The uncertainties are such that the alpha half-life so calculated is probably correct within a factor of two or three. The alpha-energy is calculated by closing a decay cycle involving beta-disintegration energies of $Pu^{241}$ and $U^{237}$ and alpha-disintegration energy of $Am^{241}$.

$Pu^{239}$   It has been mentioned in the text that there is unassigned electromagnetic radiation associated with $Pu^{239}$ and that the possibility exists that the measured alpha particle is not the ground state transition. This phenomenon would be like that which has been established for $U^{235}$. If this proves to be the case for $Pu^{239}$ the decay energy will be greater than that shown in Figure 1.

$Pu^{234}$   The degree of alpha-branching of this 8-hr. period is not accurately known because of difficulties in assessing the counting efficiency of the radiation from the electron capture process. The tentative branching ratio adopted for the present is 0.03, making the alpha half-life 10 days. The energy has been revised slightly to 6.15 Mev.[49]

<u>Pu$^{232}$</u>  The alpha-branching of this isotope has been estimated as 20% or possibly less from the alpha-activity and x-ray activity.[49]  The energy given is also only approximate since the alpha-group falls in the same energy range with its decay product U$^{228}$ which itself is not accurately known.

<u>Np$^{235}$</u>  The listed[3] alpha-branching of ~0.1% has been revised to a value of ~0.0059%.[50]

<u>Np$^{233}$</u>  This is a new neptunium isotope prepared by the irradiation of U$^{235}$ and U$^{233}$ with high energy deuterons.[51]  It decays predominantly by electron capture with a half-life of 35 min. but also shows a 5.53 Mev alpha-particle in low abundance.  As is the case with all of the nuclei decaying by electron capture without daughter activities which can be measured precisely, it is possible at present only to estimate the branching and alpha half-life.  The tentative value for the alpha-branching is ~10$^{-3}$%.

<u>U$^{235}$</u>  The main alpha group (4.396 Mev) apparently does not involve the ground state transition since there is found a 0.162 Mev gamma ray in high abundance,[3] and the decay energy shown in Figure 1 includes the gamma ray.

The abundance of the ground state transition was not known precisely since until recently it had not been observed.  On the basis of an apparent 20% discrepancy between the number of 4.396 Mev particles relative to those of U$^{238}$[52] and the number that should be present (if there were one per disintegration) according-ing to mass spectrographic analysis[53] and the yield of the actinium series products it was suggested tentatively[52] that 20% of the transitions result in a higher energy alpha-group which is covered up by the U$^{234}$ alpha particles.  Recently this laboratory obtained a sample of highly separated U$^{235*}$ and we were able to resolve a longer range alpha-group occurring in 10 $\pm$ 1% abundance and at an energy which is 160 Kev higher than the main group within the experimental uncertainty of about

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* We are indebted to Dr. C. E. Larson and other members of Carbon and Carbide Chemicals Corporation, Oak Ridge, Tennessee for making available to us the U$^{235}$ used in this study.

20 Kev. The ability to resolve this alpha-group makes it possible to determine the specific activity of $U^{235}$ directly on the highly separated material and this may well cause a change in the accepted half-life for $U^{235}$. However, rather than to attempt to correct existing data we shall retain the old value (7.07x10$^8$ yr) until these measurements can be made accurately.

$\underline{U^{233}}$ This nuclide like $Pu^{239}$ has associated with it a considerable level of electromagnetic radiation although only a single alpha-group has been identified. It is possible therefore that the observed alpha-group does not represent the ground state transition. Since there is no direct determination of a decay scheme, the measured alpha-energy is used for Figure 1.

$\underline{U^{231}}$ The alpha-energy of this 4.2-day electron capture nuclide cannot yet be measured because it cannot be prepared free of overwhelming levels of alpha-activity of $U^{230}$ and $U^{232}$. However, the extent of alpha branching can be measured by isolating the decay products $Th^{227}$ (RdAc) and $Pa^{231}$. The alpha half-life was calculated from this as 200 years.[53] The alpha-energy was estimated as 5.6 Mev both from Figure 1 and also by closing a decay cycle ($U^{231}$, $Pa^{231}$, $Ac^{227}$, $Th^{227}$) in which the electron capture decay energy for $U^{231}$ was estimated as 0.5 Mev by the relation of Thompson.[29]

$\underline{Pa^{231}}$ There is good evidence that the two well established groups of alpha particles used in this paper themselves possess fine structure and that the high energy group has a component of 5.04 Mev[54] which is about 30 Kev greater than what has been taken to be the ground state transition. This makes little difference in the energy plot of Figure 1 but would have the important effect best visualized from Figure 8 of making the ground state transition more highly forbidden that as shown. Another minor consequence of this change would be its effect on the $U^{231}$ alpha-energy since it was estimated by closing a decay cycle which includes $Pa^{231}$ alpha decay. However, other uncertainties in the calculation overshadow the slight increase in alpha-energy for $U^{231}$ that would be entailed.

$\underline{Pa^{230}}$  The degree of alpha-branching of this 17-day activity has been estimated[55] by obtaining the ratio of the $U^{230}$ and $Ac^{226}$ which grow from it assuming that the 17-day half-life is substantially that of the electron capture process rather than that of $\beta^-$-emission. The yields correspond to an alpha half-life of 1400 years which is plotted in Figure 8 against the alpha-energy (5.5 Mev) which was interpolated from Figure 1.

$\underline{Th^{230}(Io)}$  Certain aspects of the fine structure of ionium ($Th^{230}$) have been worked out and warrant incorporation into these correlations. The alpha-particle spectrum has been more carefully examined and seems to consist of at least two groups of energy 4.68 and 4.51 Mev[56] in good agreement with the known gamma ray of 68 Kev[57,58]. The ratio of the 4.68 Mev to 4.61 Mev groups is estimated by Feather[35] as $\sim 4{:}1$. Rosenblum, Valadares and Vial[56] also obtained some evidence for a group in low abundance of energy which could correspond to a transition 170 Kev above the ground state, which gamma ray has probably been observed.[57,58] However, this group is not shown in Figure 6 and Table 3 because its abundance is too poorly known.

$\underline{Th^{228}(RdTh)}$  Some newer values for the abundances of the two alpha-groups have been given as 0.72 and 0.28 for the respective group energies 5.423 and 5.338.[59]

$\underline{Th^{224}}$  The measured alpha-energy (7.20 Mev) is in keeping with a short half-life but it has not yet been measured. This is also true of some other short lived nuclides such as $Ac^{222}$, $Ra^{220}$, $Fr^{218}$, $Em^{216}$ and $Po^{211}$.

$\underline{Ac^{228}(MsTh_2)}$  A measurement of the alpha particle from rare branching has been reported[60] giving 4.54 Mev for the alpha-particle energy, a value which agrees with the predictions according to Figure 1. However, for this energy and according to Figure 8, $Ac^{228}$ should have an alpha half-life of $10^6$ years or more making the alpha-branching only 1 in $10^9$. It is extremely doubtful that such low alpha-branching could be observed in this particular case.

$Ac^{227}$   The existence of two alpha-groups one of the ~350 Kev below the main group[61] has not been confirmed[62,63] so for the present it will be assumed that $Ac^{227}$ has a single alpha-group of 4.95 Mev with the reservation that finer measurements may show this group to have closely lying fine structure. The question of whether or not the alpha particle observed represents the ground state transition remains at present unresolved.

$Ra^{226}$   There is a well recognized gamma ray of $Ra^{226}$ of about 0.19 Mev.[64,65] Stahel[65] found the conversion electrons in abundance of about 5 per 100 alpha-disintegrations and estimated an additional few percent unconverted gammas. Rosenblum and Perrin[66] observed the corresponding short range alpha-group and remarked that the abundance was of the proper magnitude to correspond with the measurements of Stahel. More recently Rosenblum[67] indicated that the abundance of the short range group is about 9% while Chang[68] gives the abundance as 1.8%. Because of the agreement between independent measurements of the gamma ray and the alpha-group by Stahel and Rosenblum respectively, the higher value is used.

$Fr^{212}$   This highly neutron deficient francium isotope (predicted β-atable francium is $Fr^{219}$) was prepared with high energy protons on thorium by Hyde, Ghiorso and Seaborg.[28] It was isolated chemically and shown to lie in the mass number range well below $Fr^{218}$ since other heavier francium isotopes which could conceivably have the observed alpha-energy and half-life would all give rise to well known decay products of the natural radioactive series. It was assigned to $Fr^{212}$ through its genetic relationship by alpha-decay with an astatine isotope thought to be $At^{208}$ and by electron capture to a new isotope of emanation assigned to $Em^{212}$ by its genetic relationship with $Po^{208}$. The measured half-life for $Fr^{212}$ is 19 min. and judging by the rate of growth of $Em^{212}$ alpha particles the alpha-branching is about 50%. The energy of the alpha particle is 6.25 Mev.

$Em^{218}$   There is a serious discrepancy in half-life for $Em^{218}$ reported by different investigators. Studier and Hyde[7] reported a half-life of 19 milliseconds for $Em^{218}$ arising as a product in the $U^{230}$ series. Walen[69], on the

other hand, claims to have identified $Em^{218}$ as a 1.3 second activity from the rare $\beta^-$-branching of $At^{218}$ which itself arose from the rare $\beta^-$-branching of $ThA^{218}$. There are several reasons for accepting the value given by Studier and Hyde, the first of which is the greatly simpler experimental situation in their method of production of $Em^{218}$. Another reason is that the energy and half-life given by Studier and Hyde fit well with the regularities of Figure 6. With regard to the 1.3-second half-life of Walen, this period is much too long for the energy given by Studier and Hyde, or conversely, if we do not accept this energy but estimate it from the 1.3 second half-life then $Em^{218}$ will have a lower energy than $An^{219}$ which is clearly out of line with the regularities of Figure 1.

$Em^{212}$ This new isotope of emanation grows from $Fr^{212}$ as mentioned and has a 6.18 Mev alpha particle.[28] The measured half-life is 23 min.[27] and since we believe that $Em^{212}$ might be beta-stable this would also be the alpha half-life.

$At^{218}$ In the review article[3] the energy of the alpha particle for $At^{218}$ found by Karlik and Bernert[34] to be 6.63 Mev was erroneously reported as 6.72 Mev.

$At^{213}$ This nuclide has never been observed and we estimate an alpha-energy (see Figure 1) following the reasoning that a nucleus in this region with 128 neutrons will have the maximum energy of all of its isotopes. This estimation was made as a prediction to emphasize the belief that this is the expected trend and that the alpha-energy of $At^{213}$ probably should not be obtained by interpolating between $At^{212}$ and $At^{214}$.

$At^{212}$ The half-life of this nuclide has been measured as 0.25 second[70] and from Figure 8 we estimate that the alpha-energy is about 7.4 Mev making allowance for nuclear type both with respect to its odd-odd composition and its decay through 126 neutrons. This energy is probably not reliable to the nearest 200 Kev at best.

It is to be expected that the alpha-energy will lie between those of $At^{211}$ and $At^{213}$ and to this extent the energy assignments given to both $At^{212}$ and $At^{213}$ are consistent.

_$At^{210}$ and lighter astatine isotopes_  Kelley and Segre[71] have for $At^{210}$ a minimum alpha-branching ratio of $10^{-4}$ which yields accordingly an alpha half-life >10yrs. From Figure 1 we may estimate the alpha-energy of $At^{210}$ as 5.4 Mev if it is assumed that it bears the same relation to $At^{211}$ as $Po^{209}$ does to $Po^{210}$. Placing this energy and half-life on Figure 9 it is seen that the alpha decay is at least several hundredfold forbidden. The fact that it is so highly forbidden is attributable partly to its having less than 126 neutrons and partly to its odd-odd configuration. It is interesting to note that (provided the energy has been guessed corectly) the prohibition of alpha-decay is greater than that of $At^{211}$ which fact may be interpreted as the effect noted for an odd-odd nucleus as compared with an odd-even one.

Ahthough alpha-emission is again observed for astatine below mass 210, isotopic assignments for the several activities cannot be made unambiguously. All of these undoubtedly decay principally by electron capture but the degree of branching has not yet even been approximated. Therefore none of these nuclides can be used in the half-life vs energy correlations. However, certain alpha-energies have been measured and isotopic assignments were made as indicated in Figure 1. It should be emphasized that the half-lives given here are the measured half-lives and not those for alpha-emission.

The assignment of a 5.5-hr period with alpha particles of 5.65 Mev energy has been tentatively made to $At^{209}$ based upon the excitation function in the irradiation of bismuth with helium ions and the probable identification of $Po^{209}$ among the decay products of the complex mixture of activities.[72] Another 5.65 Mev alpha particle decaying with a half-life of 1.7 hr has been assigned to $At^{208}$, and was produced through alpha-decay of an activity thought to be

$Fr^{212}$.[28] It was assigned to $At^{208}$ principally on the basis of the appearance of $Po^{208}$ at the rate corresponding to the decay of a 1.8 hr. parent. It may be mentioned that by spallation reactions of bismuth another apparent astatine parent of $Po^{208}$ is produced which decays with a longer half-life than 1.8 hrs. and has no observable alpha-radiation.[72] It is possible that there are isomers of $At^{208}$ which show up in different abundances by the two modes of formation.

Through excitation function measurements a 5.76 Mev alpha particle decaying with 1.8-hr. half-life has been assigned to $At^{207}$.[72] Two other probable astatine isotopes are assigned to $At^{205}$ and $At^{204}$ somewhat arbitrarily although the mass number range is defined by excitation experiments. The two activities are respectively a 25 min. period of 5.9 Mev alpha-energy and a 10 min. period with a 6.1 Mev alpha particle.[72]

$Po^{209}$ This new isotope of polonium has a 200-year half-life for alpha-decay estimated from yield considerations[71] and the alpha particle energy has been revised slightly as 4.90 Mev.[73] The electron capture branching is not known accurately but was estimated from the amount of L x-rays as a maximum of 1 in 10.[24] The electron capture half-life would be accordingly greater than 2000 years.

$Po^{205}$ and lighter polonium isotopes As in the case of light astatine isotopes there is little accurate information on the alpha half-lives of these species and the isotopic assignments are not certain. The energies are sufficiently well known and the isotopic assignments are well enough defined for use in Figure 1 to show the trend expected in this region. The presently accepted decay properties of $Po^{205}$ are a measured half-life of 1.5 hr. with a 5.2 Mev alpha particle.[24] The 5.35 Mev alpha particle with 4-hour half-life[3] has been reassigned to $Po^{204}$[24] while the 40 min., 5.56 Mev alpha-emitter is retained at $Po^{203}$. In each case the degree of alpha branching is not known.

$Bi^{208}(?)$  In a previous communication[10] a 5.0 Mev alpha particle found by Howland and Perlman(unpublished) was attributed to $Bi^{208}$. This alpha particle appeared as a very weak activity in the bismuth fraction of pile neutron irradiated bismuth and was thought to arise from an n,2n reaction. This assignment was not very attractive since we might expect $Bi^{208}$ to have an alpha-energy not greater and perhaps less than that of $Bi^{209}$. The interesting possibility is being explored[74] that the 5.0 Mev alpha-group belongs to a metastable state of $Bi^{210}$(RaE) highly forbidden toward isomeric transition and beta decay. Partial but inconclusive evidence for this assignment has been obtained.

$Bi^{201}$  The 60-min. activity with alpha particles of 5.15 Mev assigned to $Bi^{200}$(3) has been reassigned to $Bi^{201}$ on the basis of its genetic relationship to 8-hr. Pb and 72-hr Tl which are tentatively assigned to mass number 201.[21]

$Bi^{199}$  This is the only one of the neutron deficient bismuth alpha-emitters for which the alpha-branching has now been estimated. Its isotopic assignment has been made[21] by observing the growth through successive electron capture processes of $Pb^{199}$ and the 7.5-hr. $Tl^{199}$. The alpha-branching calculated from the yields of the $Tl^{199}$ and the observed alpha-emission rate is $1.7 \times 10^{-3}\%$ which gives an alpha half-life of 3 yrs.

$Bi^{197}$  This 2-min. activity assigned to $Bi^{197}$ is used in these correlations in terms of its minimum alpha half-life by assuming that the measured half-life is controlled by the alpha-decay process.

$Au^{190}$  This alpha-emitter[3] has been more positively identified as an isotope of gold and by measuring the other radiation decaying with a 5-min. half-life the alpha-branching was estimated as $10^{-2}\%$.[23]

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Figure 1

Alpha-Energy vs. Mass Number

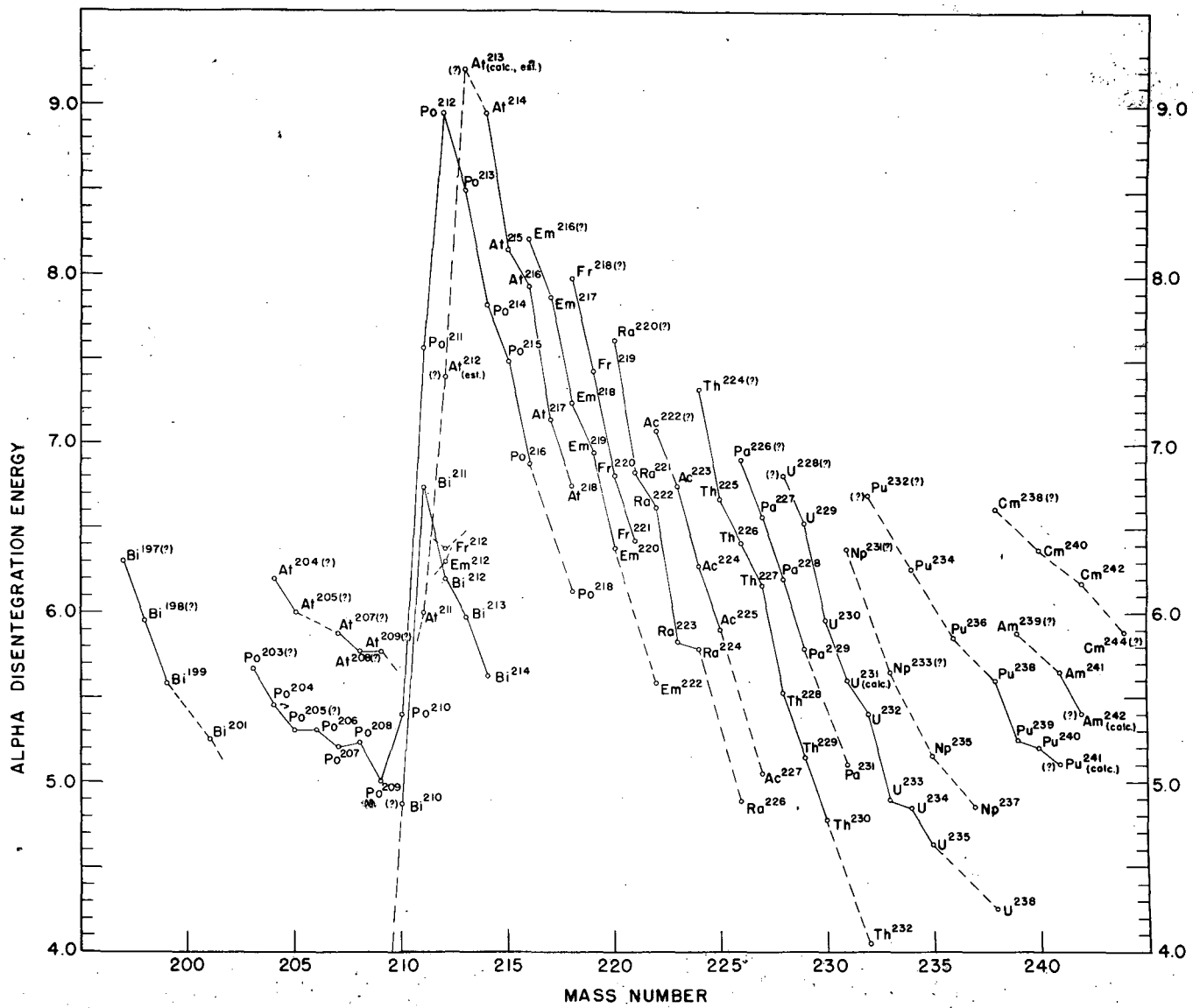Relationships of the Heavy Nuclides

Alpha disintegration energy versus mass number for isotopes of Bi, Po, At, Em, Fr, Ra, Ac, Th, Pa, U, Np, Pu, Cm.

OZ.623

Figure 2

Parabolic sections of energy surface

showing increase in alpha-energy with
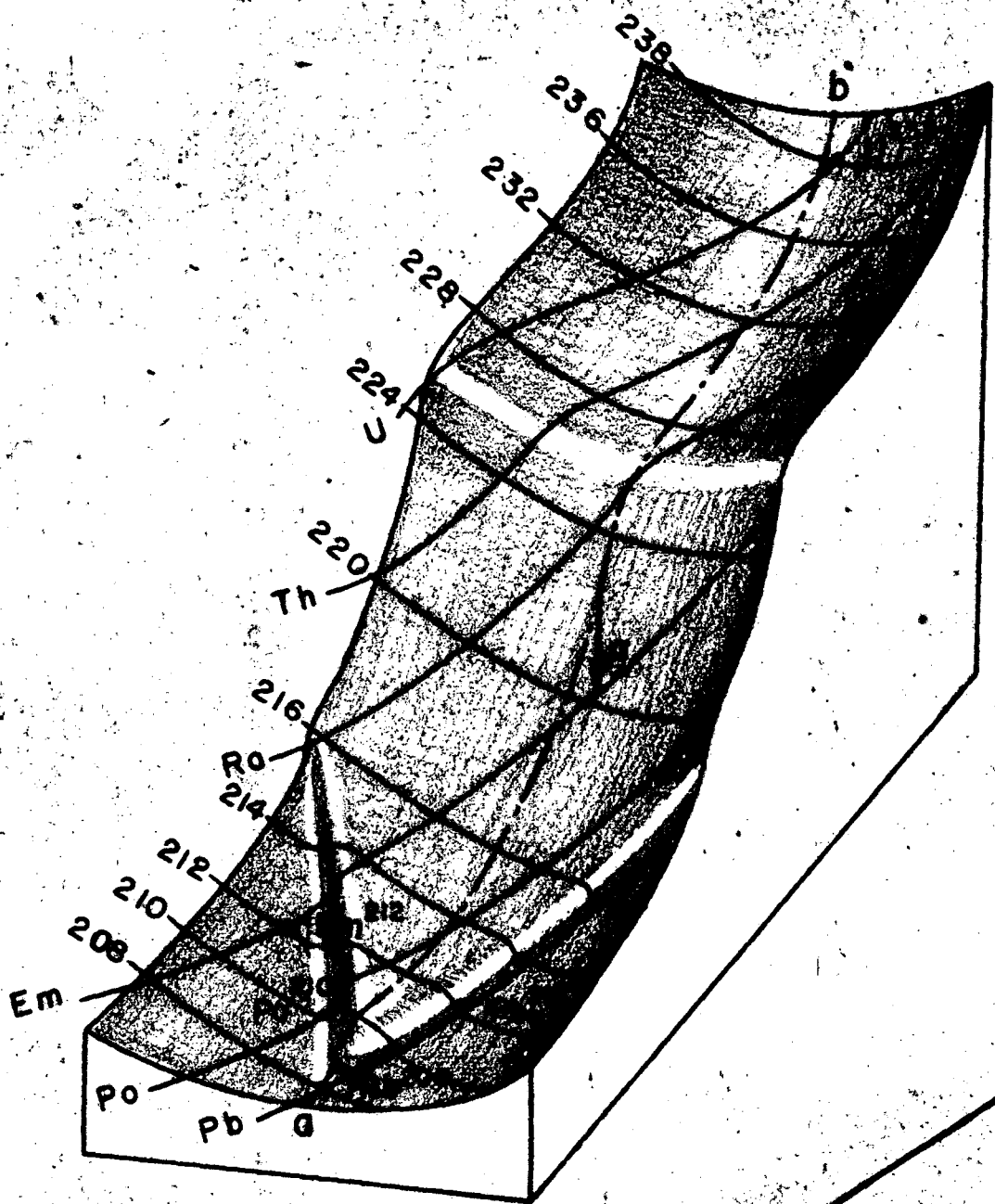
mass number decrease

ENERGY →

INCREASING ATOMIC NUMBER →
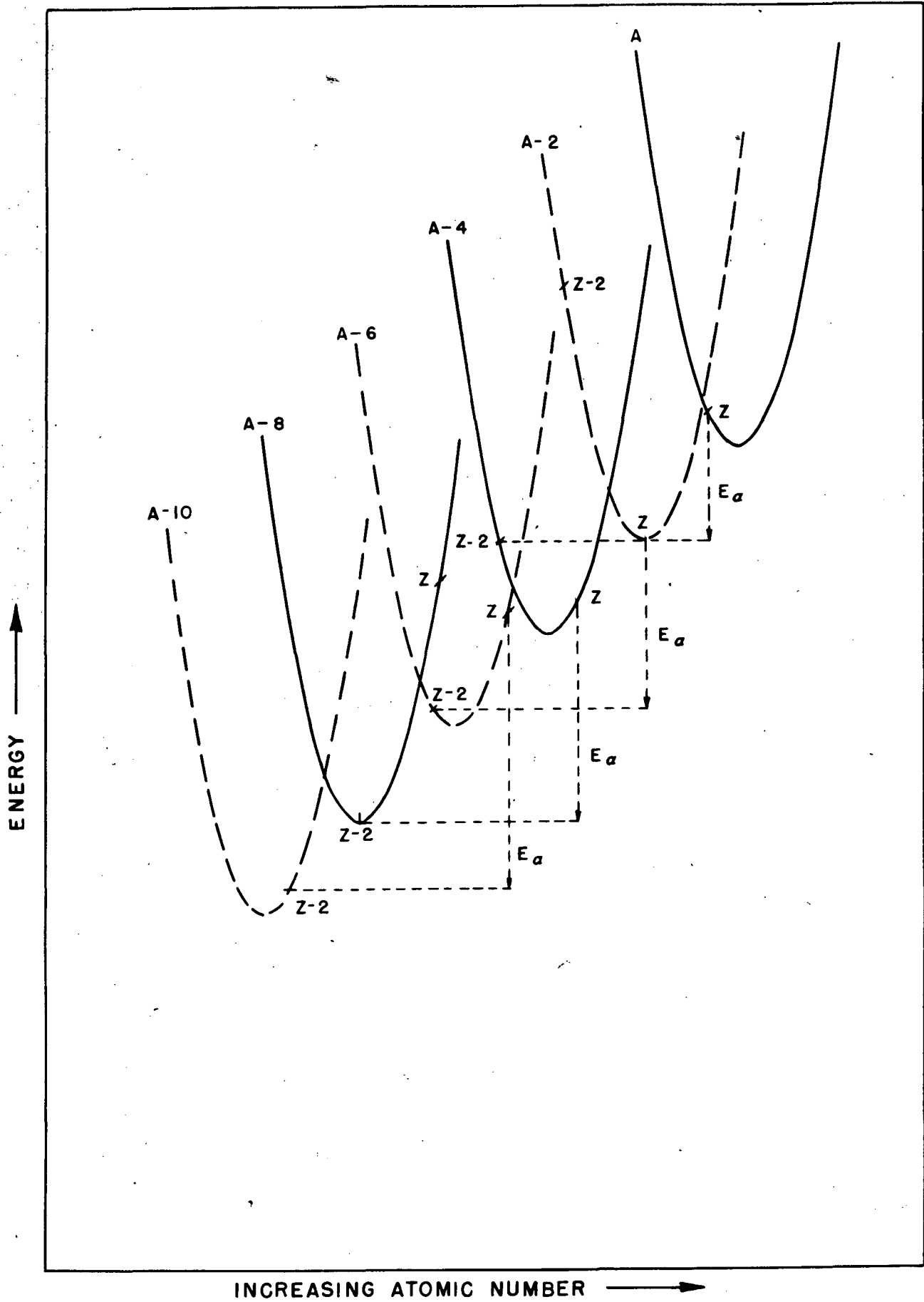
Figure 3

Energy Surface in the

Heavy Element Region

Figure 4

Alpha-Energies Trends of Heavy Nuclides

(Heavy solid lines join nuclides of comparable

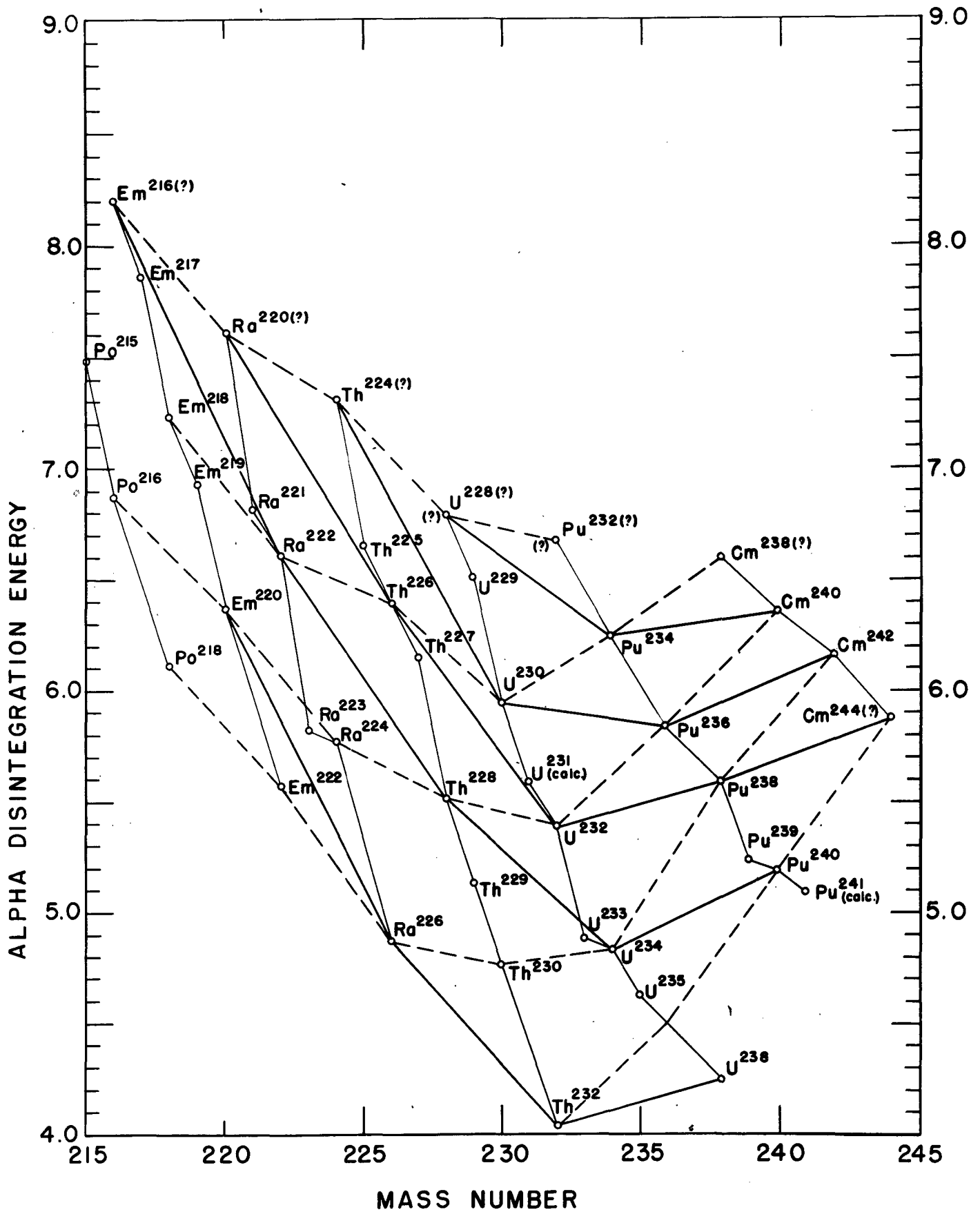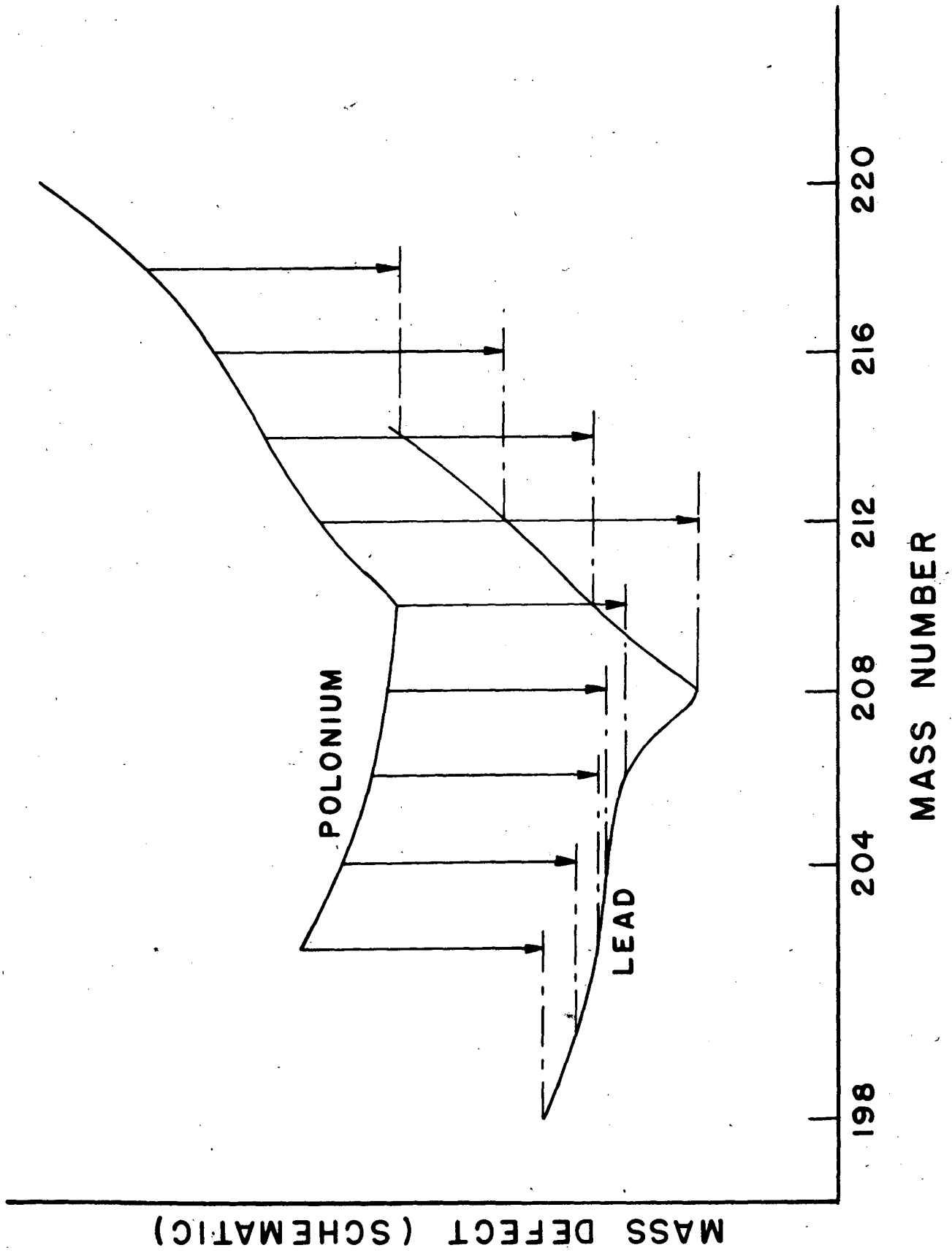beta stability;  broken lines join nuclides

along alpha-decay chain)

OZ 618

Figure 5

Schematic sections of energy surface at Z = 84 and

Z = 82 illustrating trend of alpha-energies of the

polonium isotopes (arrows drawn at polonium isotope

mass number;   length of arrows proportional to

alpha energy)

POLONIUM

LEAD

MASS NUMBER

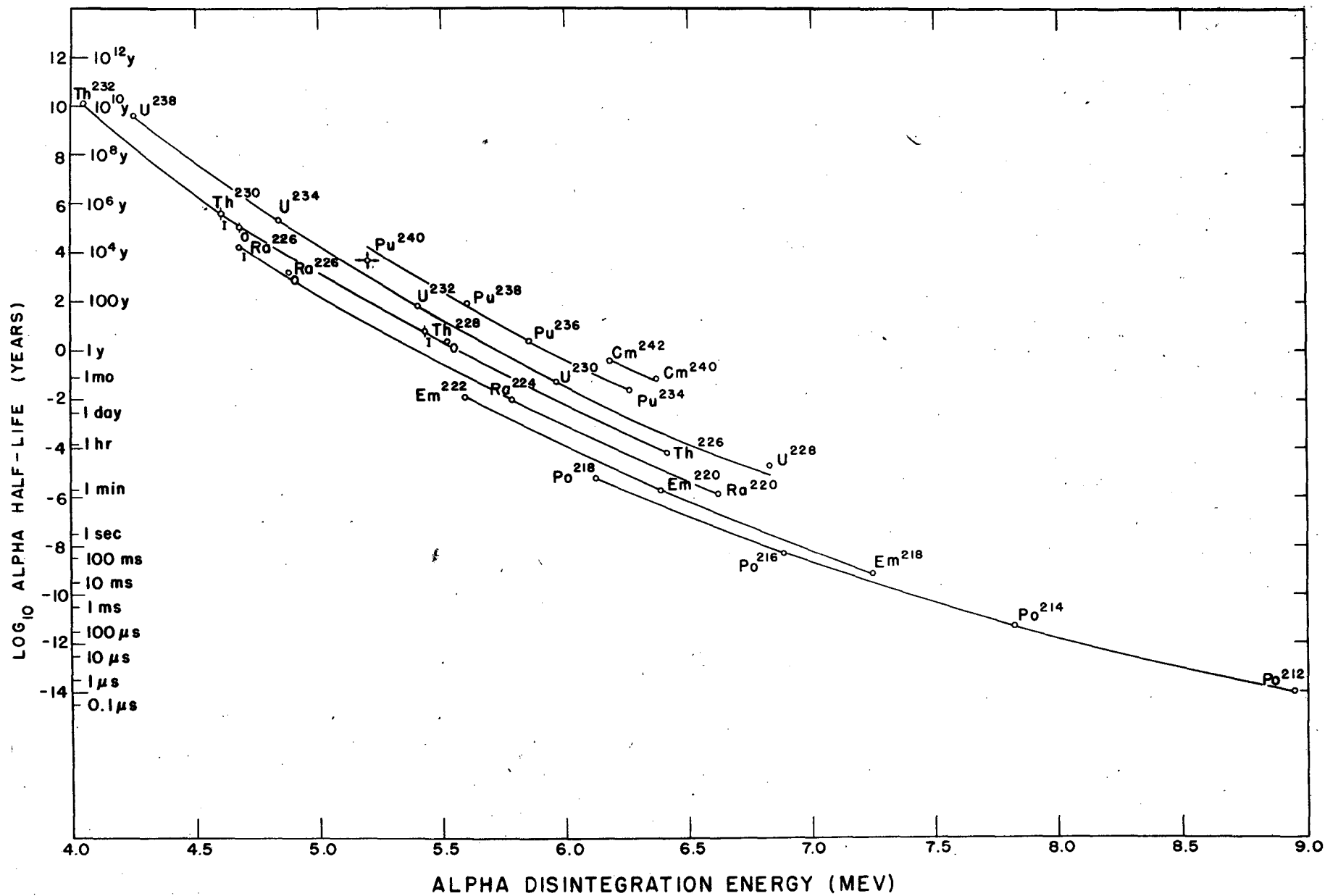MASS DEFECT (SCHEMATIC)

198    204    208    212    216    220

Figure 6

Half-life vs. Energy Relationship

for the Even-Even Nuclides

(Roman numerals indicate short-range groups

in fine structure and "0" the ground state transition)

Figure 7

Half-life vs. Energy Relationship of

the Even-Odd Nuclides

(Roman numerals indicate short-range groups

in fine structure and "0" the ground state transition)

Figure: Plot of $\text{LOG}_{10}$ ALPHA HALF-LIFE (YEARS) versus ALPHA DISINTEGRATION ENERGY (MEV).

Y-axis labels: $10^{12}$ y, $10^{10}$ y, $10^{8}$ y, $10^{6}$ y, $10^{4}$ y, 100 y, 1 y, 1 mo, 1 day, 1 hr, 1 min, 1 sec, 100 ms, 10 ms, 1 ms, 100 $\mu$s, 10 $\mu$s, 1 $\mu$s, 0.1 $\mu$s

Data points labeled: $U^{235}$, $U^{235}$, $U^{233}$, $Pu^{241}$, $Th^{229}$, $Th^{229}$, $Pu^{239}$, $Th^{229}$, $U^{231}$, $Ra^{223}$, $Th^{227}$, $Th^{227}$, $Ra^{223}$, $U^{229}$, $Th^{225}$, $Em^{249}$, $Ra^{221}$, $Em^{219}$, $Po^{215}$, $Em^{217}$, $Po^{213}$

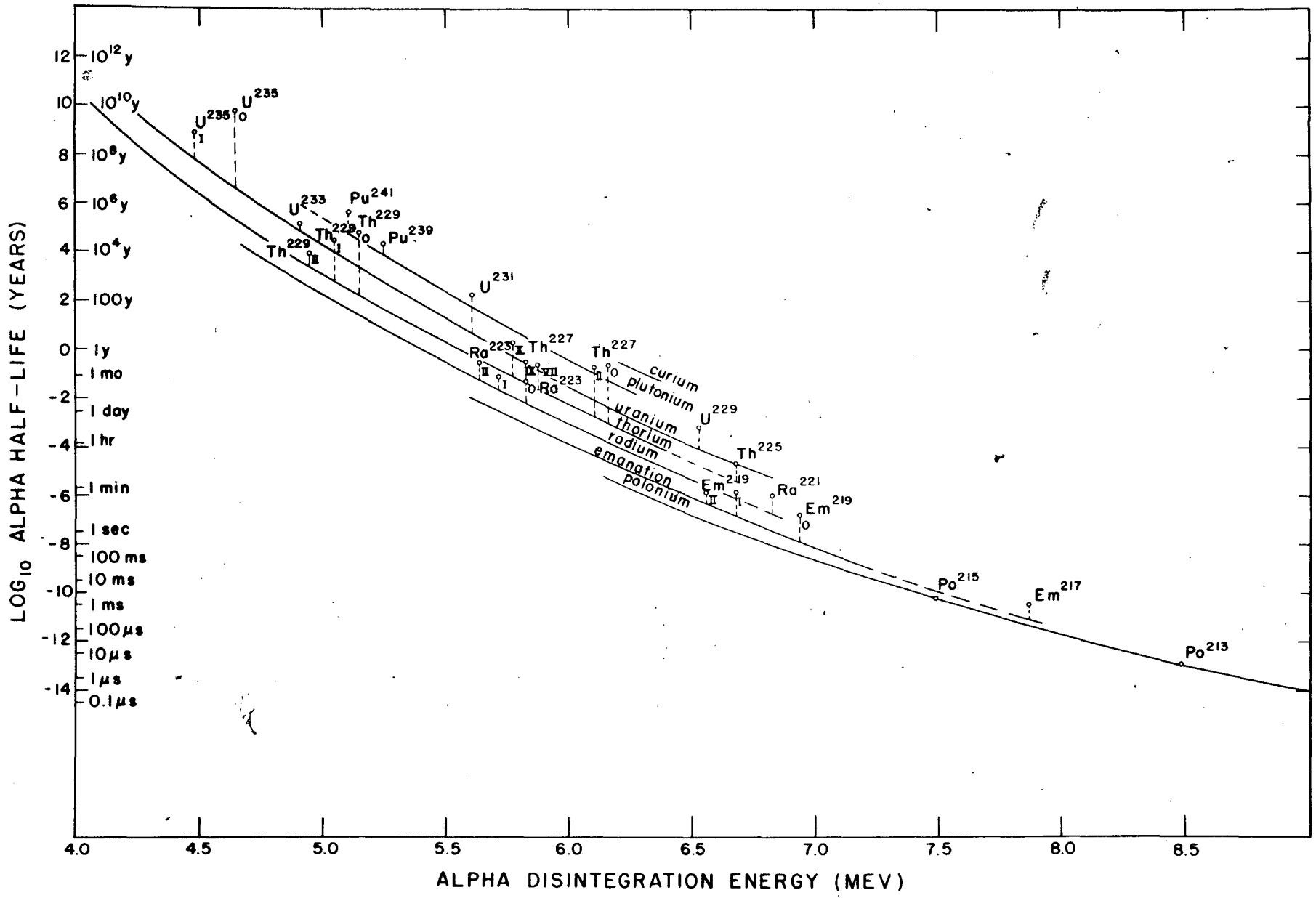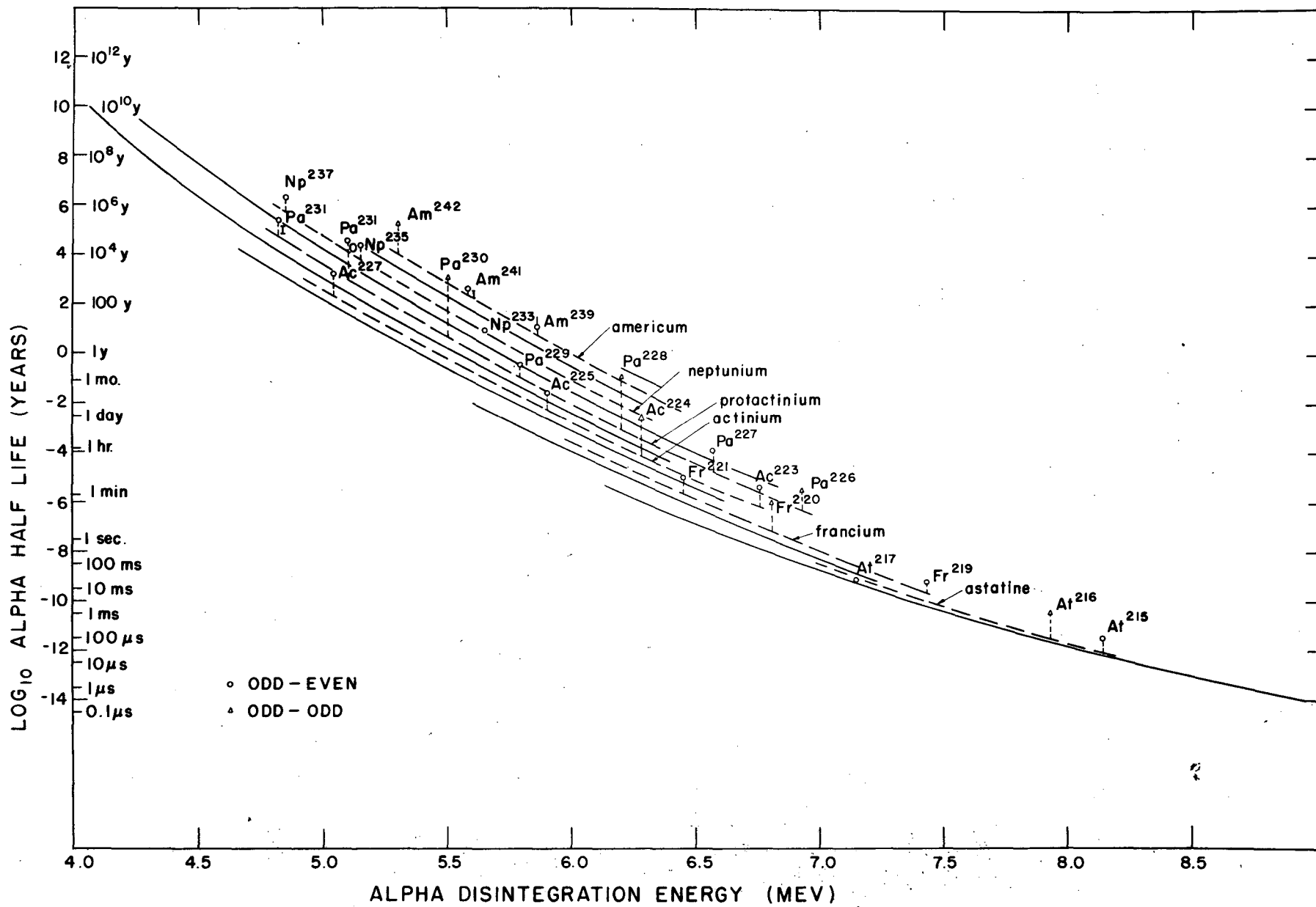Curve labels: curium, plutonium, uranium, thorium, radium, emanation, polonium

OZ 620

Figure 8

Half-life vs. Energy Relationship

for the Odd-Even and Odd-Odd Nuclides

(Roman numerals indicate short-range groups

in fine structure and "0" the ground state transition)

Figure 9

Half-life vs. Energy Relationship of

the Specially Forbidden Nuclear Types

(Roman numerals indicate short-range groups

in fine structure and "0" the ground state transition)

ALPHA DISINTEGRATION ENERGY (MEV)

OZ 619