

Systematics of Alpha-Radioactivity

I. PERLMAN, A. GHIORSO, AND G. T. SEABORG

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

(Received September 26, 1949)

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 percent. 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 $r = 1.48A^{1/3} \cdot 10^{-13}$ cm seems to fit the data for the even-even nuclei outside of the region of 126 neutrons better than more complex functions.

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 development 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

¹ H. Geiger and J. M. Nuttall, *Phil. Mag.* **22**, 613 (1911); **23**, 439 (1912).

² G. Gamow, *Zeits. f. Physik* **51**, 204 (1928); *Structure of Atomic Nuclei and Nuclear Transformations* (Oxford University Press, London, 1937).

E. U. Condon and R. W. Gurney, *Phys. Rev.* **33**, 127 (1929); *Nature* **122**, 439 (1928).

H. A. Bethe, *Revs. Mod. Phys.* **9**, 161 (1937).

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 introduction 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 considerable 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

³ G. T. Seaborg and I. Perlman, *Revs. Mod. Phys.* **20**, 585 (1948).

⁴ References for this work as well as other work not specifically cataloged here will be found in the "Table of Isotopes" which is reference 3.

preparation of U^{233} in quantity has added 7 more alpha-emitters as members of the $4n+1$ radioactive family⁶ while 4 and 18 new species, respectively, were added by the discovery of the U^{230} collateral series⁷ 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 predictions 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

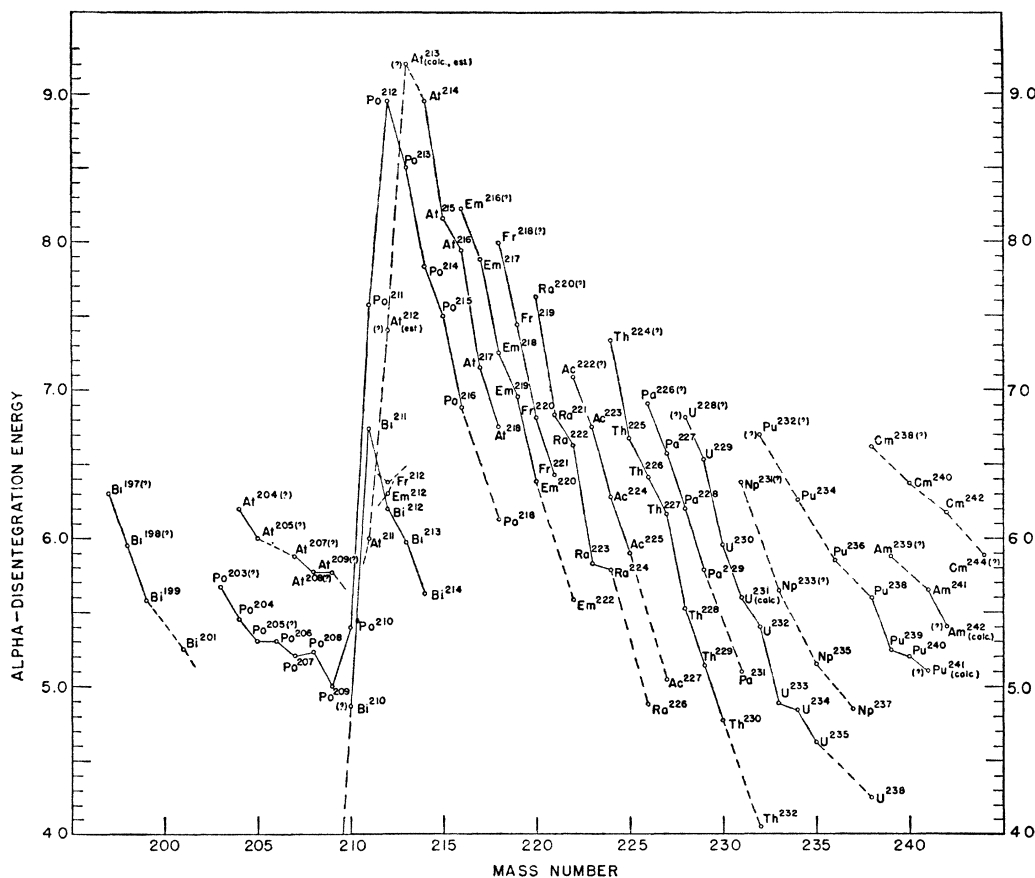


FIG. 1. Alpha-energy *vs.* mass number relationships of the heavy nuclides.

⁵ Hagemann, Katzin, Studier, Ghiorso, and Seaborg, Phys. Rev. **72**, 252 (1947).

⁶ English, Cranshaw, Demers, Harvey, Hincks, Jelley, and May, Phys. Rev. **72**, 253 (1947).

⁷ M. H. Studier and E. K. Hyde, Phys. Rev. **74**, 591 (1948).

⁸ Ghiorso, Meinke, and Seaborg, Phys. Rev. **74**, 695 (1948); Meinke, Ghiorso, and Seaborg, Phys. Rev. **75**, 314 (1949).

⁹ Templeton, Howland, and Perlman, Phys. Rev. **72**, 527 (1947); D. H. Templeton and I. Perlman, Phys. Rev. **73**, 1211 (1948); for others, see references in the Appendix.

¹⁰ Perlman, Ghiorso, and Seaborg, Phys. Rev. **74**, 1730 (1948); **75**, 1096 (1949).

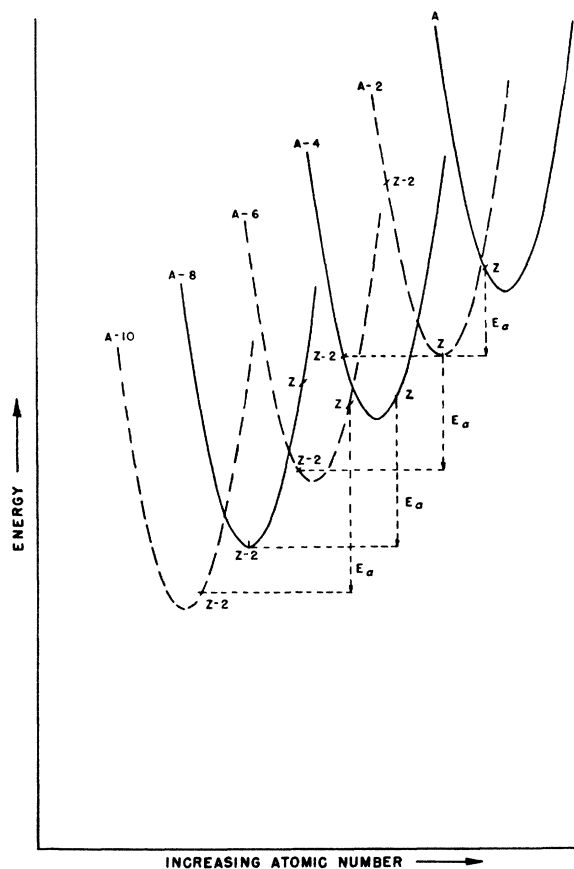


FIG. 2. Parabolic sections of energy surface showing increase in alpha-energy with mass number decrease.

against and which parameter is chosen as the basis for joining points.

One of the early methods of treating data was that of Fournier¹¹ 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. Schintlmeister¹² 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 Berthelot¹³ and more recently by Karlik¹⁴ using methods somewhat similar to those mentioned.

Another method of correlating data on alpha-decay used recently by Wapstra¹⁵ and Glueckauf¹⁶ 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.* $A-Z$. Contour 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 in idealized form for A , $A-2$, $A-4$, etc. The parabolic form of these sections at constant A was deduced by Bethe and Bacher¹⁷ 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 Fig. 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 Fig. 2

¹¹ G. Fournier, *Comptes Rendus* 184, 878 (1927); K. Fajans, *Radioelements and Isotopes* (McGraw-Hill Book Company, Inc., New York, 1931), Chapter I.

¹² J. Schintlmeister, *Wien. Chem. Zeits.* 46, 106 (1943).

¹³ A. Berthelot, *J. de phys. et rad.* VIII, 3, 17 (1942).

¹⁴ B. Karlik, *Acta Phys. Austriaca* 2, 182 (1948).

¹⁵ A. H. Wapstra, *Nature* 161, 529 (1948).

¹⁶ E. Glueckauf, *Proc. Phys. Soc.* 61, 25 (1948).

* 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."

¹⁷ H. A. Bethe and R. F. Bacher, *Revs. Mod. Phys.* 8, 82 (1936) (pp. 104 and 165).

¹⁸ N. Bohr and J. A. Wheeler, *Phys. Rev.* 56, 426 (1939).

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 Fig. 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. Kohman¹⁹ has developed an expression for alpha-energy in terms of the Bohr-Wheeler mass equation¹⁸ 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.

The above explanation with Fig. 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 Fig. 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 Fig. 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 Fig. 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 Fig. 3 by the arrow labeled " α ". 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.

The manner in which these inferences were made may be clarified by reference to Fig. 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 Fig. 1. The contours shown in heavy lines on Fig. 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 Fig. 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 Fig. 4 may best be visualized by examining the model in Fig. 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 farther one proceeds up the

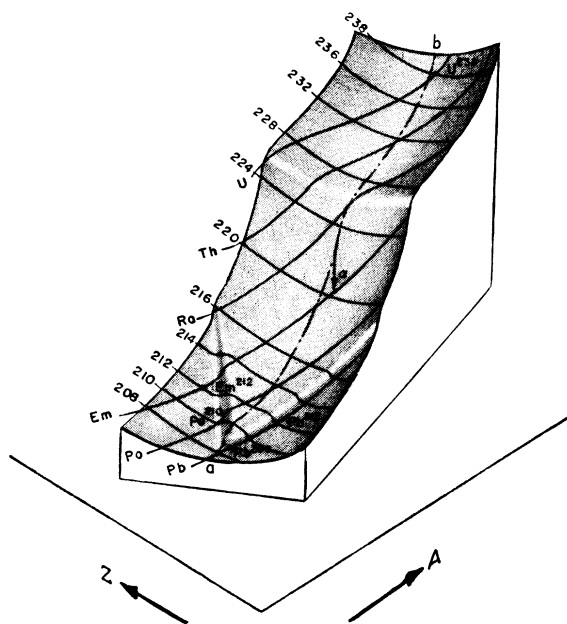


FIG. 3. Energy surface in the heavy element region.

¹⁹ T. P. Kohman, Phys. Rev. 76, 448 (1949).

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

slope and they will become lower the farther 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 Fig. 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

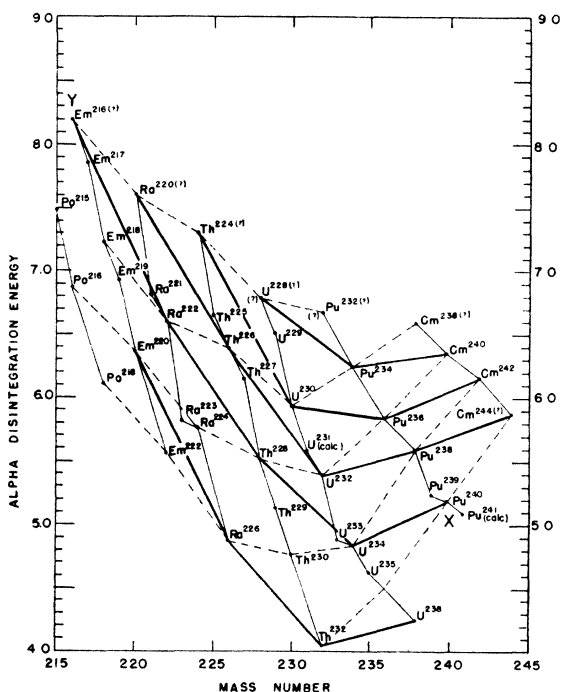


FIG. 4. Alpha-energies trends of heavy nuclides. (Heavy solid lines join nuclides of comparable beta-stability; broken lines join nuclides along alpha-decay chain.)

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 ten- 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 con-

ditions, 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 Fig. 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 has 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 Fig. 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 Fig. 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 Fig. 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 Fig. 3. The changes in slope of the curves of Fig. 5

²⁰ Bi^{202} and Bi^{206} decay by electron capture with half-lives of about 2 hr. and 14 days, respectively (D. G. Karraker and D. H. Templeton, unpublished data); Bi^{203} decays by electron capture with 12-hr. half-life (H. M. Neumann and I. Perlman, unpublished).

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 Fig. 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

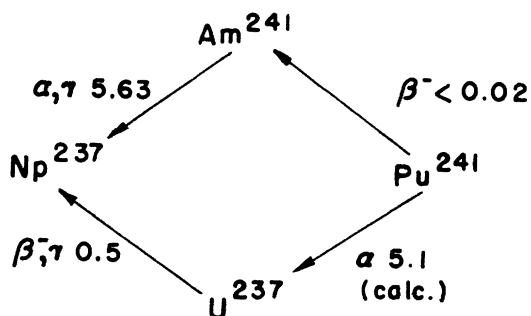
It is inherent in most of the methods used for determining alpha-energies that fairly accurate values are obtained. Almost none of the energies shown should be in error by more than 100 keV, and most of them are known with considerably better accuracy. It will be seen in the final section of this paper 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 that arise from other sources. There are a number of cases of uncertain assignment of mass number and these are indicated in Fig. 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 Fig. 1 are accompanied by question marks preceding the symbol as in the case of Pu²³² 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.

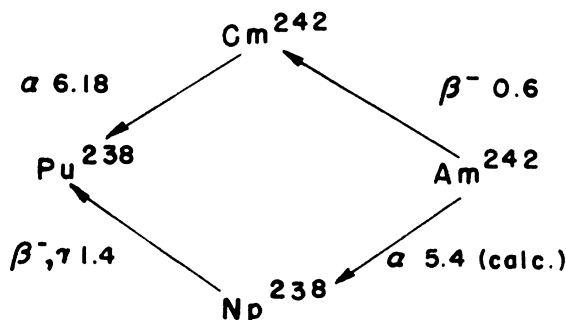
Beyond the uncertainties already mentioned it should be realized that total decay energy is the property which should be compared and there has been a tacit assumption that the alpha-energies shown represent ground state transitions. Where this condition is known not to apply, corrections have been made by addition of the gamma-ray energy as in the cases of U²³⁵ and Am²⁴¹ (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, Pu²³⁹ and U²³³, both of which would seem to be out of line in Fig. 1, have associated with them considerable gamma-radiation³ 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, Bi²¹⁰(RaE), Pu²⁴¹ and Am²⁴², 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 Feather²² 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 Po²¹⁰, it was possible to calculate the alpha-energy of RaE. The following diagrams designate the data used in calculating the alpha-energies for Pu²⁴¹ and Am²⁴² (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 (Fig. 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

²¹ H. M. Neumann and I. Perlman (unpublished).

²² E. Broda and N. Feather, Proc. Roy. Soc. (London) A190, 20 (1947).

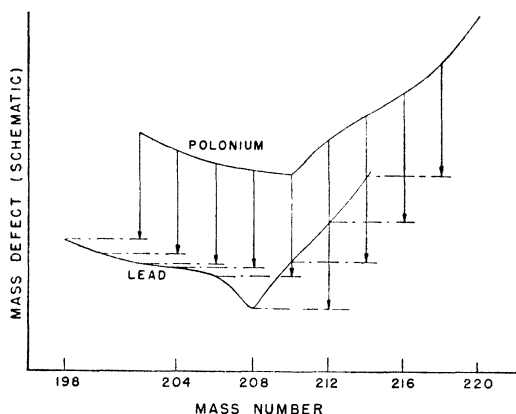


FIG. 5. Schematic sections of energy surface at $Z=84$ and $Z=82$ illustrating trend of alpha-energies of the polonium isotopes (arrows drawn at polonium isotope mass number; length of arrows proportional to alpha-energy).

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 interruption 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 Fig. 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 laboratory²³ 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 experiments²⁴ 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 Fig. 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 Fig. 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 Fig. 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 Fig. 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 Fig. 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

²³ Thompson, Ghiorso, Rasmussen, and Seaborg, *Phys. Rev.* **76**, 1406 (1949).

²⁴ D. G. Karraker and D. H. Templeton, unpublished data.

²⁵ W. Elsasser, *J. de phys. et rad.* **5**, 625 (1934).

²⁶ M. G. Mayer, *Phys. Rev.* **74**, 235 (1948).

²⁷ Ghiorso, Meinke, and Seaborg, *Phys. Rev.* **76**, 1414 (1949).

²⁸ Hyde, Ghiorso, and Seaborg (unpublished).

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

Methods and Examples

It is apparent that one may use the correlations of Fig. 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 discernible 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.

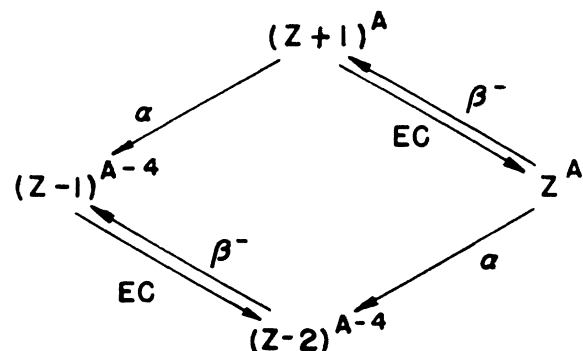
There are a number of reasons for wishing to know whether a nucleus is beta-stable or unstable irrespective of its alpha-decay properties. The beta-stable nuclei in this region are entirely analogous to the stable nuclei lower in the periodic table and their pattern constitutes a source of information 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 I) 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 requirements 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.

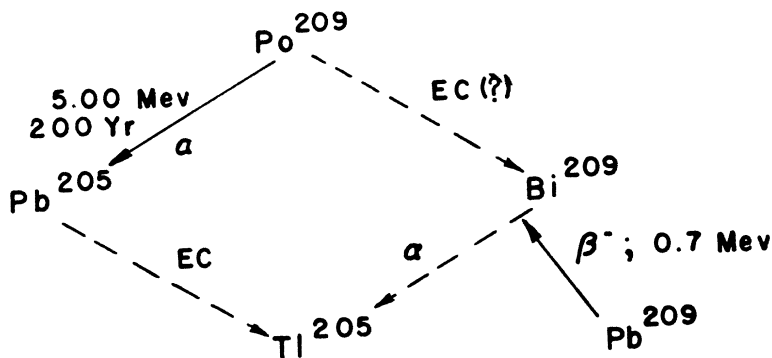
The methods of making these predictions vary in kind and reliability. Alpha-energies may be read off a plot such as Fig. 1 by interpolation or extrapolation bearing in mind the sharp changes which occur in the nuclei near closed shell configurations. For example, one may be fairly certain that the alpha-energy of Em^{221} lies between those of Em^{220} and Em^{222} and has an energy of 6.0 ± 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 Fig. 1) in an analogous fashion to the pairs $^{\text{A}}\text{Bi}^{211} - ^{\text{A}}\text{Bi}^{212}$ and $^{\text{A}}\text{Po}^{212} - ^{\text{A}}\text{Po}^{213}$.

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. Sometimes 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 allows one to set an upper limit of 10 percent 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 $\text{Po}^{209} \rightarrow \text{Bi}^{209}$ 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^{238} family which has a 3.3 hr. half-life and 0.7 Mev decay energy. This half-life and energy correspond to an allowed transition which Feather and Richardson³⁰ 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 Mayer³¹ 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 Hammack³³ 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 Fig. 1 although this is only a prediction. It will be noted that At^{214} has almost the same alpha-energy as $\text{ThC}'(\text{Po}^{212})$ 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 β^- -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 $\text{Po}^{213} - \text{At}^{213}$ 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

²⁹ S. G. Thompson, Phys. Rev. **76**, 319 (1949).

³⁰ N. Feather and H. O. W. Richardson, Proc. Phys. Soc. **61**, 452 (1948).

³¹ M. G. Mayer, Phys. Rev. **75**, 1969 (1949).

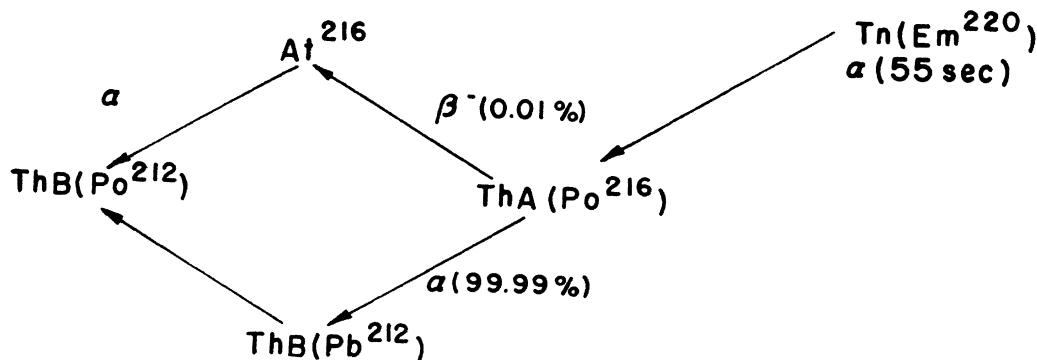
³² L. W. Nordheim, Phys. Rev. **75**, 1894 (1949).

³³ E. Feenberg and K. C. Hammack, Phys. Rev. **75**, 1877 (1949).

for each element in that isotope which decays to a product with 126 neutrons, in this case $\text{At}^{213} \rightarrow \text{Bi}^{209}$. To this estimated decay energy of At^{213} must be added the electron capture energy of At^{213} or the β^- -energy of Po^{213} must be subtracted, whichever applies.

At^{216} is reported by Karlik and Bernert³⁴ to arise in

the thorium decay series through β^- -branching in the decay of $\text{ThA}(\text{Po}^{216})$. 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 percent branching in ThA decay according to the following scheme:



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 a 1.15-Mev excited state of $\text{Bi}^{212}(\text{ThC})$ 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 Fig. 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 series⁸ 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 $\text{RdTh}(\text{Th}^{228})$ from electron capture branching of Pa^{228} , of $\text{ThX}(\text{Ra}^{224})$ from electron capture branching of Ac^{224} and $\text{ThC}'(\text{Po}^{212})$ from the β^- -decay of $\text{ThC}(\text{Bi}^{212})$. All in all, it appears to be highly unlikely that $\text{ThA}(\text{Po}^{216})$ 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 Feather³⁵ 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 I 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 Mukherjee³⁶ listing types of instability, differs in some instances from Table I 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 laboratory⁸ on the alpha-energy of At^{216} gave 7.79 Mev which when used with the alpha-energy of $\text{Po}^{216}(\text{ThA})$ and the decay energy of $\text{Pb}^{212}(\text{ThB})$ allows

³⁴ B. Karlik and T. Bernert, *Zeits. f. Physik* **123**, 51 (1944).

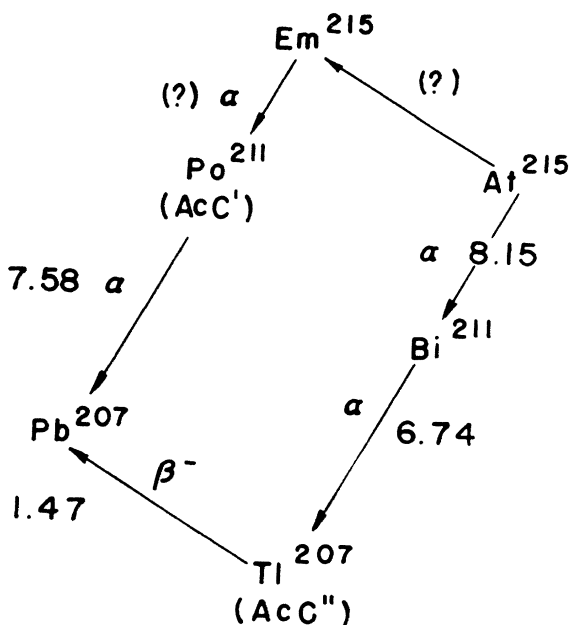
³⁵ N. Feather, *Nucleonics* **5**, 22 (1949).

³⁶ S. Biswas and A. Mukherjee, *Ind. J. Phys.* **22**, 80 (1948).

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 measured⁸ alpha-energy of At^{214} , it is possible to show that $\text{Po}^{214}(\text{RaC}')$ 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 Fig. 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 $\text{At}^{213}-\text{Po}^{213}$. 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 Fig. 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 Fig. 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 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 promethium (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 ${}_{39}\text{Cb}^{93}$ and ${}_{45}\text{Rh}^{101}$. 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 niobium (41) or technetium (43).

Promethium (element 61) is in an analogous position with respect to neutron number 82. Here there is a long list of stable nuclei, namely, ${}_{54}\text{Xe}^{136}$, ${}_{56}\text{Ba}^{138}$, ${}_{57}\text{La}^{139}$, ${}_{58}\text{Ce}^{140}$, ${}_{59}\text{Pr}^{141}$, ${}_{60}\text{Nd}^{142}$, ${}_{62}\text{Sm}^{144}$. 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, promethium, through reasoning similar to that used for technetium.

¶ Note added in proof.—In an article which came to our attention since the preparation of this manuscript, A. Broniewski (Comptes Rendus 228, 916 (1949)) had already noted the occurrence of the missing elements 43 and 61 near positions of closed neutron shells.

TABLE I. Beta-stability in the heavy elements.*

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

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 $_{82}\text{Pb}^{208}$, $_{83}\text{Bi}^{209}$, and $_{84}\text{Po}^{210}$. The interesting possibility has already been considered that $_{86}\text{Em}^{212}$ 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 $\text{Ra}^{223}(\text{AcX})$. 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 $\text{Em}^{219}(\text{An})$ 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 Fig. 1 (see Table III) 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 $\text{Em}^{219}(\text{An})$ 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 β^- -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 Fig. 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 II 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.

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 Fig. 6 allow one to extrapolate to element 98 and

TABLE II. 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^{246} —(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)	

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 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} or both on this picture but the best direct evidence to date places the assignment at Sm^{152} .³⁷

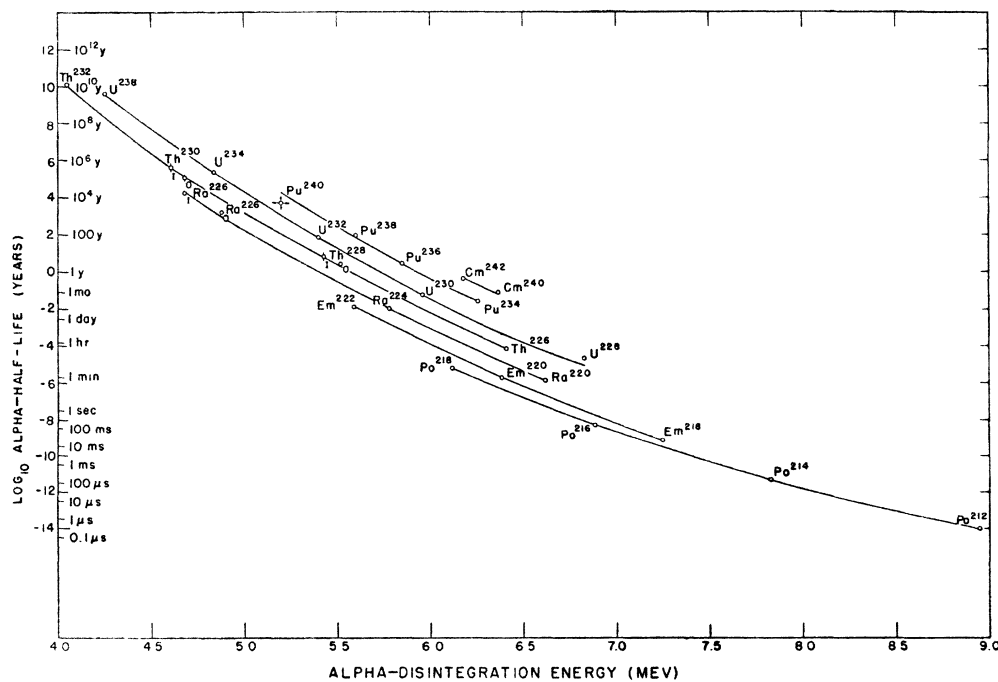


FIG. 6. Half-life vs. energy relationship for the even-even nuclides. (Roman numerals indicate short-range groups in fine structure and "Q" the ground state transition.)

³⁷ A. J. Dempster, *Phys. Rev.* **73**, 1125 (1948).

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 formula² 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 percent 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 III 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 III are entered on a series of diagrams (Figs. 6-8, 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 forbidden in their alpha-decay and the last column in Table III 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 Fig. 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 (Fig. 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 Fig. 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 shrinkage in nuclear radius in 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 Berthelot³⁸ and Biswas³⁹ 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 Fig. 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

³⁸ A. Berthelot, J. de phys. et rad. VIII 3, 52 (1942).

³⁹ S. Biswas, Ind. J. Phys. 23, 51 (1949).

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 Fig. 6. That is to say, the partial alpha-half-lives for the two groups of each of the nuclides Ra²²⁶, Th²³⁰(Io) and Th²²⁸(RdTh) are what would be expected for the respective energies of the

TABLE III. 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 yr.		~6000 yr.	
Pu ²³⁸	5.60		92 yr.		92 yr.	
Pu ²³⁶	5.85		2.7 yr.		2.7 yr.	
Pu ²³⁴	6.26		8.5 hr.	~0.03	~10 days	
U ²³⁸	4.25		4.51×10 ⁹ yr.		4.51×10 ⁹ yr.	
U ²³⁴	4.84		2.35×10 ⁶ yr.		2.35×10 ⁶ yr.	
U ²³²	5.40		70 yr.		70 yr.	
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		1.39×10 ¹⁰ yr.		1.39×10 ¹⁰ yr.	
Th ²³⁰	0 4.76	0.80	8.0×10 ⁴ yr.		1.0×10 ⁶ yr.	
	I 4.69	0.20			4.0×10 ⁵ yr.	
Th ²²⁸	0 5.52	0.70	1.90 yr.		2.64 yr.	
	I 5.43	0.28			6.79 yr.	
Th ²²⁶	6.41		30.9 min.		30.9 min.	
Ra ²²⁶	0 4.88	0.91	1622 yr.		1780 yr.	
	I 4.68	0.09			1.8×10 ⁴ yr.	
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.	
Po ²¹⁴	7.83		1.5×10 ⁻⁴ sec.		1.5×10 ⁻⁴ sec.	
Po ²¹²	8.95		3.0×10 ⁻⁷ sec.		3.0×10 ⁻⁷ sec.	
Even-odd species						
Pu ²⁴¹	5.1 (calc.)		~10 yr.	2×10 ⁻⁵	5×10 ⁵ yr.	9
Pu ²³⁹	5.24				2.4×10 ⁴ yr.	4
U ²³⁵	0 4.64	0.10	7.07×10 ⁸ yr.		7.1×10 ⁹ yr.	1000
	I 4.48	0.90			7.7×10 ⁸ yr.	13
U ²³³	4.90		1.6×10 ⁶ yr.		1.6×10 ⁶ yr.	2
U ²³¹	5.6 (est.)		4.2 days	6×10 ⁻⁵	200 yr.	40
U ²²⁹	6.53		58 min.	0.2	5 hr.	9
Th ²²⁹	0 5.14	0.1	7000 yr.		7×10 ⁴ yr.	350
	I 5.04	0.2			3.5×10 ⁴ yr.	45
	II 4.94	0.7			10 ⁴ yr.	3
Th ²²⁷	0 6.16	0.20	18.6 days		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	0 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	0 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(?)
Po ²¹⁶	7.50		1.83×10 ⁻³ sec.	~1(?)	1.8×10 ⁻³ sec.	1
Po ²¹³	8.49		4×10 ⁻⁶ sec.		4×10 ⁻⁶ sec.	1

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

TABLE III. (Cont.)

Nucleus	Alpha-energy	Abundance of α -group	Measured half-life	Alpha-branching ratio	Alpha-half-life	Departure* factor
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		2.2×10 ⁶ yr.		2.2×10 ⁶ yr.	4
Np ²³⁶	5.15		435 days	5×10 ⁻⁵	2.5×10 ⁴ yr.	4(?)
Np ²³³	5.65		35 min.	10 ⁻⁵	10 yr.	1(?)
Pa ²³¹	0 5.10	0.87	3.43×10 ⁴ yr.		3.95×10 ⁴ yr.	40
	I 4.82	0.13			2.64×10 ⁶ 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 ²²⁶	5.90				10.0 days	5
Ac ²²³	6.76		2.2 min.	~1(?)	2.2 min.	7
Fr ²²¹	6.45		4.8 min.	~1(?)	4.8 min.	5
Fr ²¹⁹	7.44		0.02 sec.		0.02 sec.	3(?)
At ²¹⁷	7.15		0.021 sec.	~1(?)	0.021 sec.	1
At ²¹⁵	8.15		10 ⁻⁴ sec.	~1(?)	10 ⁻⁴ sec.	4(?)
Odd-odd species						
Am ²⁴²	5.4 (calc.)				~2×10 ⁶ yr.	20(?)
Pa ²³⁰	5.5 (est.)		17.7 days	3.5×10 ⁻⁶	1400 yr.	250(?)
Pa ²²⁸	6.20		22 hr.	~0.02	~50 days	150(?)
Pa ²²⁶	6.93		1.7 min.	~1(?)	1.7 min.	6
Ac ²²⁴	6.28		2.9 hr.	0.1	29 hr.	40
Fr ²²⁰	6.81		27.5 sec.	~1(?)	27.5 sec.	13
At ²¹⁶	7.94		10 ⁻³ sec.	~1(?)	10 ⁻³ sec.	10
Specially forbidden species (Bismuth isotopes and others with 126 neutrons or less)						
Bi ²¹⁴	0 5.61	0.45	19.7 min.	4×10 ⁻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. (11.3 days)	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	~3×10 ⁻⁷	5×10 ⁴ yr.	10 ⁴ (?)
Bi ¹⁹⁹	5.58		25 min.	~2×10 ⁻⁶	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.	0.5	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 yr.	150
Po ²⁰⁸	5.24		3 yr.		3 yr.	30
Po ²⁰⁷	5.2		5.7 hr.	~10 ⁻⁴	7 yr.	50(?)
Po ²⁰⁶	5.3		9 days	0.1	90 days	6(?)
Po ²⁰⁴	5.45		4 hr.	~10 ⁻⁴	5 yr.	1000
Au ^{<190}	5.3		4.3 min.	10 ⁻⁴	1 mo.	400(?)

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 Fig. 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 Fig. 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 III) 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 Th²²⁹ in which comparison with the curve of Fig. 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 three-fold forbidden. All of the other cases of fine structure are qualitatively similar to that of Th²²⁹ 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 $\text{Th}^{227}(\text{RdAc})$, $\text{Ra}^{223}(\text{AcX})$, and $\text{Em}^{219}(\text{An})$ 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 ± 1 percent 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 tenfold 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 Figs. 1 and 7. From Fig. 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 Fig. 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 Fig. 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 Fig. 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 Quantitative Treatment of Alpha Decay). 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 Fig. 8 in which the same reference lines for the even-even nuclei are shown as in Figs. 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 III.

For the odd-even species there is reason to doubt that the departures from the reference curves as shown in Table III and Fig. 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 III 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 Fig. 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 Figs. 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

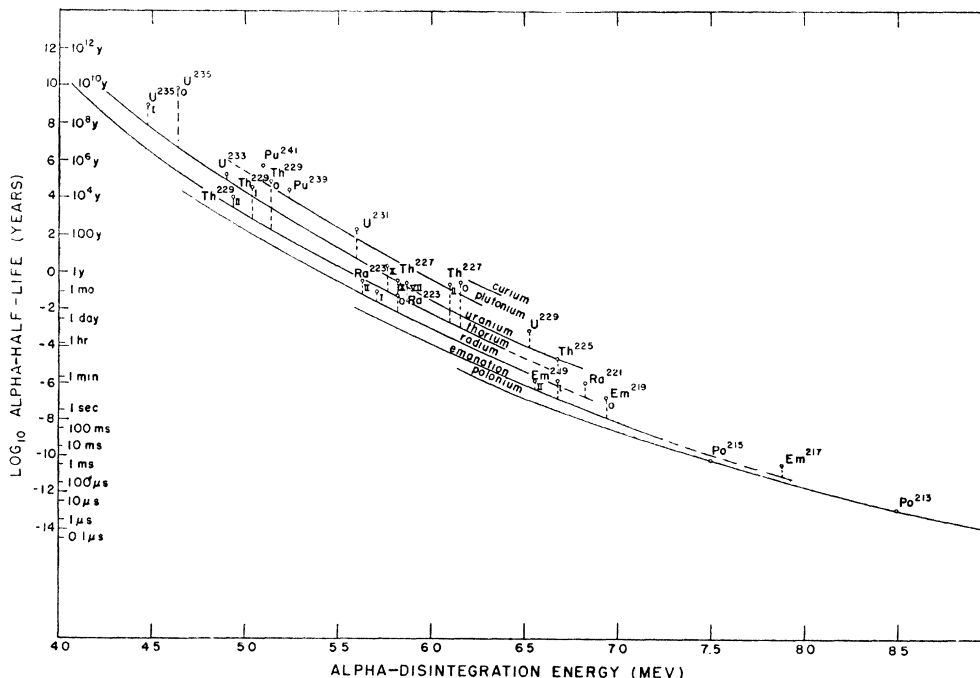


FIG. 7. Half-life vs. energy relationship of the even-odd nuclides. (Roman numerals indicate short-range groups in fine structure and "0" the ground state transition.)

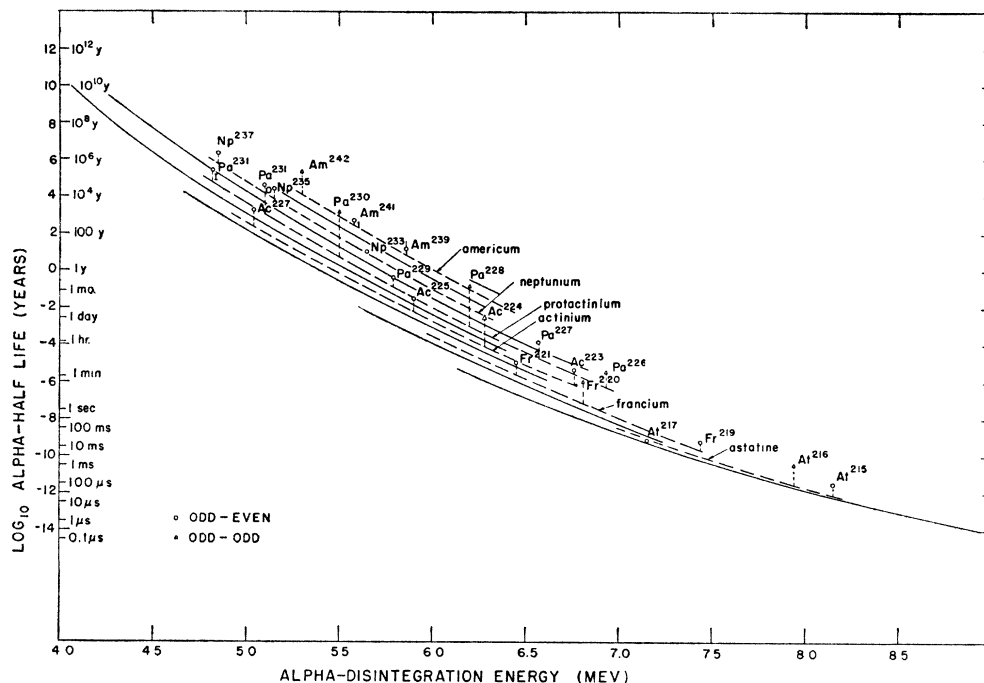


FIG. 8. Half-life *vs.* energy relationship for the odd-even and odd-odd nuclides. (Roman numerals indicate short-range groups in fine structure and "0" the ground state transition.)

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 obtained⁴⁰ 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 λ 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 antiparallel 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 III and Figs. 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

⁴⁰ O. E. Andersen and H. E. White, Phys. Rev. **71**, 911 (1947).

⁴¹ H. Schüler and H. Gollnow, Naturwiss. **22**, 511 (1934).

these species. It is worth while to attempt to correlate such abnormalities in nuclear dimensions as evidenced by alpha-decay properties with the newer 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 ascribed 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.

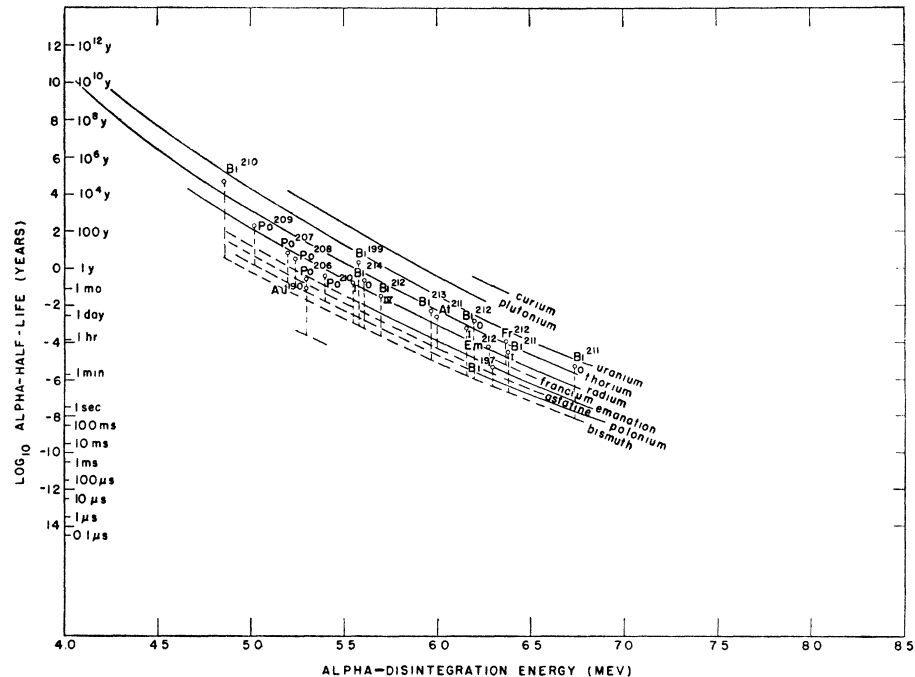
As already mentioned all of the naturally occurring isotopes of bismuth show very highly forbidden alpha-decay ranging from several hundred-fold to several thousand-fold above the baseline curve. Part of this may now be attributed to odd nucleons in bismuth isotopes, but there is still a considerable 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 percent could account for a 50-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 percent 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 percent 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 III must be considered only as rough approximations. The two-minute period assigned to Bi^{197} is only about tenfold 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

FIG. 9. Half-life vs. energy relationship of the specially forbidden nuclear types. (Roman numerals indicate short-range groups in fine structure and "Q" the ground state transition.)



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 forbidden 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 Fig. 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 Fig. 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 implication of the abnormally long half-lives in both cases is that there are discontinuities 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 128 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 $\text{Po}^{211}(\text{AcC}')$ and leads to the prediction of that for At^{212} as shown in Fig. 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 tenfold greater than that predicted presumably from the Geiger-Nuttall relations.⁴²

This discussion of the effect of 126 neutrons on the

⁴² International Radium Standards Commission Report of 1930, *Rev. Mod. Phys.* 3, 427 (1931).

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 Fig. 6 are reproduced rather faithfully by the single body theory of alpha-emission as has already been indicated by Biswas³⁹ 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 $r = 1.48A^{1/3} \cdot 10^{-13}$ cm, will fit 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, $r = 1.48A^{1/3} \cdot 10^{-13}$ cm. In about a dozen such calculations for nuclides from emanation to curium, the mean deviation encountered was about 1 percent 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 measurements 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

*** We wish to thank Mr. T. J. Ypsilantis for making most of the computations referred to here.

⁴³ H. A. Bethe, *Rev. Mod. Phys.* 9, 163 (1937).

⁴⁴ E. Amaldi and B. N. Cacciapuoti, *Phys. Rev.* 71, 739 (1947).

has long been apparent in the decay of Po^{210} and possibly 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 configuration 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 percent, 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 percent.

It may be mentioned that further dissociation of factors which affect 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 fortuitously, Em^{212} is less forbidden than is Po^{210} and Po^{208} .

This work was performed at the Radiation Laboratory, University of California, Berkeley, California, under the auspices of the AEC.

APPENDIX

Notes and Comments on Individual Activities

Most of the data used in this paper may be found in the compilation "Table of Isotopes."⁷³ 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 listed³ this is an alpha-emitter of 6.50 Mev and 2.5-hr. half-life. It is also surely unstable with respect to electron capture (see Table I) 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 Fig. 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 shown⁴⁵ to be less than 20 percent 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. Thompson⁴⁶ 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 Fig. 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 III 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 measurements⁴⁷ 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 Fig. 1.

Am^{239}

The listed³ alpha-branching of ~ 0.1 percent has been redetermined⁴⁸ and a better value 0.01 percent has been used here in determining the alpha-half-life.

Pu^{241}

The alpha-half-life indicated in Table III 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 pos-

⁴⁵ K. Street, Jr. and G. T. Seaborg, unpublished data.

⁴⁶ S. G. Thompson, Ph.D. Thesis, University of California (1948) "Nuclear and Chemical Properties of Americium and Curium."

⁴⁷ Goeckermann, Robinson, and Stover, unpublished data.

⁴⁸ R. A. James and K. Street, Jr., unpublished data.

sibility 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 Fig. 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

Pu^{232}

The alpha-branching of this isotope has been estimated as 20 percent 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.

Np^{235}

The listed³ alpha-branching of ~ 0.1 percent has been revised to a value of ~ 0.005 percent.⁵⁰

Np^{233}

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 $\sim 10^{-3}$ percent.

U^{235}

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 Fig. 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 percent discrepancy between the number of 4.396 Mev particles relative to those of U^{233} ,⁵² and the number that should be present (if there were one per disintegration) according to mass spectrographic analysis and the yield of the actinium series products it was suggested tentatively⁵² that 20 percent 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 ± 1 percent abundance and at an energy which is 180 kev higher than the main group with an experimental uncertainty of about 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 (7.07×10^8 yr.) until these measurements can be made accurately.

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

⁴⁹ D. A. Orth and G. T. Seaborg, unpublished data.

⁵⁰ R. A. James, unpublished data.

⁵¹ Magnussen, Thompson, and Seaborg, unpublished data.

⁵² Clark, Spencer-Palmer, and Woodward, British Atomic Energy Project Report, BR-522 (1944).

† 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.

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 Fig. 1.

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 Fig. 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

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 Mev⁵⁴ 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 Fig. 1 but would have the important effect best visualized from Fig. 8 of making the ground state transition more highly forbidden than 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.

Pa^{230}

The degree of alpha-branching of this 17-day activity has been estimated⁵⁵ 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 β -emission. The yields correspond to an alpha-half-life of 1400 years which is plotted in Fig. 8 against the alpha-energy (5.5 Mev) which was interpolated from Fig. 1.

$Th^{230}(Io)$

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 4.61 Mev⁵⁶ 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 Feather⁵⁵ as $\sim 4:1$. Rosenblum, Valadares, and Vial⁵⁶ 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 Fig. 6 and Table III because its abundance is too poorly known.

$Th^{228}(RdTh)$

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

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} .

⁵³ Crane, Ghiorso, and Perlman, unpublished data.

⁵⁴ S. Rosenblum and E. Cotton, *Comptes Rendus* **226**, 171 (1948).

⁵⁵ W. W. Meinke, and G. T. Seaborg, unpublished data.

⁵⁶ Rosenblum, Valadares, and Vial, *Comptes Rendus* **227**, 1088 (1948).

⁵⁷ A. G. Ward, *Proc. Camb. Phil. Soc.* **35**, 322 (1939).

⁵⁸ I. Curie, *Comptes Rendus* **227**, 1225 (1948).

⁵⁹ Rosenblum, Valadares, and Perey, *Comptes Rendus* **228**, 385 (1949).

$\text{Ac}^{228}(\text{MsTh}_2)$

A measurement of the alpha-particle from rare branching has been reported⁶⁰ giving 4.54 Mev for the alpha-particle energy, a value which agrees with the predictions according to Fig. 1. However, for this energy and according to Fig. 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 group⁶¹ 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} Stahel⁶⁵ found the conversion electrons in abundance of about 5 per 100 alpha-disintegrations and estimated an additional few percent unconverted gammas. Rosenblum and Perrin⁶⁶ 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 Rosenblum⁶⁷ indicated that the abundance of the short-range group is about 9 percent while Chang⁶⁸ gives the abundance as 1.8 percent. 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 -stable 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 percent. 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 Hyde⁷ 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 β^- -branching of At^{218} which itself arose from the rare β^- -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

⁶⁰ G. Guében, Ann. soc. sci. Bruxelles **B52**, 60 (1932); **B53**, 115 (1933).

⁶¹ R. G. Gregoire and M. Perey, Comptes Rendus **225**, 733 (1947).

⁶² L. Vigneron, Comptes Rendus **226**, 715 (1948).

⁶³ Ghiorso, Hollander, and Perlman, U. S. Atomic Energy Commission Document, AEC-D-2232 (1948).

⁶⁴ O. Hahn and L. Meitner, Zeits. f. Physik **26**, 161 (1924).

⁶⁵ E. Stahel, Comptes Rendus **194**, 608 (1932).

⁶⁶ S. Rosenblum and J. Perrin, Comptes Rendus **195**, 317 (1932).

⁶⁷ S. Rosenblum, Nucleonics **4**, 38 (1949).

⁶⁸ W. Y. Chang, Phys. Rev. **70**, 632 (1946).

⁶⁹ R. Walen, Comptes Rendus **227**, 1090 (1948).

energy and half-life given by Studier and Hyde fit well with the regularities of Fig. 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 Fig. 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 article³ the energy of the alpha-particle for At^{218} found by Karlik and Bernert³⁴ 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 Fig. 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 second⁷⁰ and from Fig. 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 Segrè⁷¹ have for At^{210} a minimum alpha-branching ratio of 10^{-4} which yields accordingly an alpha-half-life > 10 years. From Fig. 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 Fig. 9 it is seen that the alpha-decay is at least several hundred-fold 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 correctly) 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.

Although 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 Fig. 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

⁷⁰ Weissbluth, Putnam, and Segrè, private communication (July 1948).

⁷¹ E. L. Kelley and E. Segrè, Phys. Rev. **75**, 999 (1949).

⁷² Barton, Ghiorso, and Perlman, unpublished data.

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 considerations⁷¹ 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 Fig. 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-life³ 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.

⁷³ A. Ghiorso, unpublished data.

$\text{Bi}^{208}(\text{?})$

In a previous communication¹⁰ 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 explored⁷⁴ 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 made²¹ 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 1.7×10^{-3} percent which gives an alpha-half-life of 3 years.

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-emitter³ 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} percent.²³

⁷⁴ Neumann, Howland, and Perlman, unpublished data.

The Beta-Spectra of Cl^{38} *

LAWRENCE M. LANGER**

Los Alamos Scientific Laboratory, Los Alamos, New Mexico

(Received September 23, 1949)

The beta-spectra of the 38-min. activity of Cl^{38} was studied in a magnetic lens spectrometer. The momentum distribution was resolved into three groups with end points of 4.81, 2.77, and 1.11 Mev. The highest energy group was found to have a shape characteristic of a once forbidden transition involving a spin change of 2 units and a change of parity.

INTRODUCTION

THE 38-min. activity of Cl^{38} has been studied by many investigators.¹ Most recent measurements with magnetic spectrometers have been made by Hole and Siegbahn² and by Watase and Itoh.³ In both cases,

* This document is based on work performed under government Contract No. W-7405-Eng-36 for the Los Alamos Scientific Laboratory of the University of California.

** Indiana University, Bloomington, Indiana.

¹ Complete references to earlier work may be found in reference 2.

² N. Hole and K. Siegbahn, *Arkiv. f. Mat., Astr. o. Fys.* **33A**, No. 9 (1946).

³ Y. Watase and J. Itoh, *Proc. Phys. Math. Soc. Japan* **30**, 626 (1939).

the beta-transition was found to be complex with at least two and probably three groups of electrons.

By means of a Fermi plot analysis, Hole and Siegbahn arrive at end points of 1.19, 2.70, and 5.2 Mev. However, because of the effect of saturation in the iron of their spectrometer, they express some uncertainty about the accuracy of the high energy group which in turn raises some doubts about the exact determination of the inner end points. They prefer, therefore, to base the value of the beta-end points upon the energy measurement of the two cascade gamma-rays which they measure as 1.60 Mev and 2.15 Mev. They assume that the energy determination of the lowest beta-group is

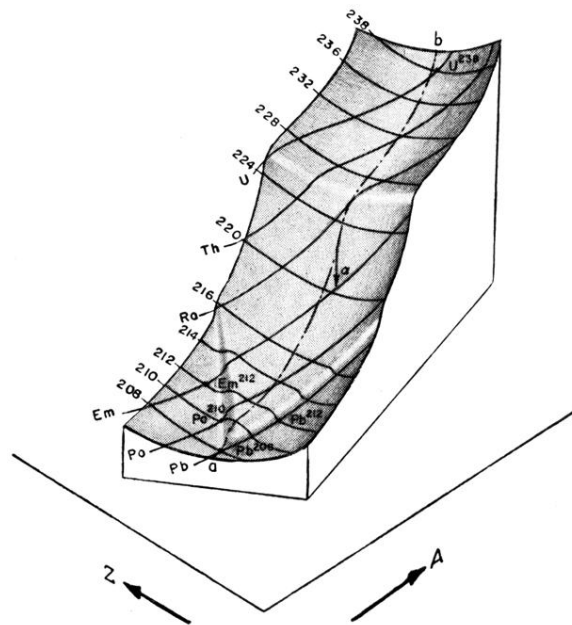


FIG. 3. Energy surface in the heavy element region.