Long-Lived Bi^{207} and Energy Levels of Pb^{207}

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The long-lived isotope of bismuth, Bi^{207}, has been prepared by deuteron irradiation of lead and from the alpha-decay of At^{211}. From the genetic relationship with At^{211}, the half-life is estimated as 50 years. The conversion electron spectrum has been determined and is discussed in terms of the energy states of Pb^{207}. Further evidence on these states has been obtained from the discovery of three rare alpha-groups in the decay of Po^{211} of energies 6.90, 6.57, and 6.34 Mev, which are compared with the main group at 7.43 Mev.

I. INTRODUCTION

THE region of naturally occurring bismuth isotopes lies in the range from Bi^{209} to Bi^{214}, but isotopes down to Bi^{198} have been produced by cyclotron irradiations.[1–3] Two isotopes, Bi^{207} and Bi^{208}, have never been reported although sought for[1,4] and have been presumed to be either long-lived or very short-lived, probably the former. The present report deals with the properties of Bi^{207} produced under more favorable conditions by two methods previously tried: the deuteron irradiation of lead[1] and the alpha-decay[4] of At^{211}.

This nucleus, Bi^{207}, decays by electron capture; and its gamma-ray spectrum is of particular interest in indicating energy levels of Pb^{207}. The lowest lying energy levels of lead isotopes seem to be widely spaced as indicated, for example, by the well-known lowest excited state of Pb^{208} at 2.62 Mev and by other data from radioactive decay processes.[5]

Other measurements of energy states of Pb^{207} have been obtained in the present studies by observation of multiple alpha-groups in the decay of Po^{211}, which is best obtained without conflicting alpha-activities by preparing its electron capture parent At^{211}. Also included in this report are some observations on the failure to find any activity from Bi^{208} which can be explained in terms of the energy states of Pb^{208}.

II. Bi^{207} FROM THE ALPHA-DECAY OF At^{211}

The 7.5-hr isotope At^{211} decays 40 percent by emission of a 5.89-Mev alpha-particle[6] and 60 percent by electron capture to give Po^{211}(AcC′), which in turn decays with a 7.43-Mev alpha-particle. Because of the short half-life of Po^{211} (estimated to be several milliseconds), it cannot be dissociated from its parent, so that At^{211} appears to decay with two alpha-groups widely separated in energy. The alpha-decay of Po^{211} results in stable Pb^{207} and the alpha-branching of At^{211} produces Bi^{207}, which should be unstable toward decay by electron capture to Pb^{207}.

Four large sources of At^{211} (each $\sim 10^9$ disintegrations/min) were prepared by bombarding four thick target samples of metallic bismuth with 38-Mev helium ions. Under these conditions roughly equivalent amounts of At^{210} and At^{211} are formed. The presence of At^{210} does not interfere with the measurements to be made. The astatine was separated from each target by volatilizing it from the molten target onto a cooled silver disk.

Two of the silver plates were used for counting purposes without further purification of the astatine. The alpha-decay of these samples showed a 7.2-hour decay (At^{211}), followed by the longer-lived Po^{210} which arises from the electron capture decay of At^{210}. The decay of the At^{210} gamma-ray was measured with a Geiger-Müller counter using a 1008-mg/cm² Pb absorber to eliminate x-rays from both At^{210} and At^{211}, and a half-life of 8.0 hr was observed. The residual activity remaining after complete decay of the astatine isotopes was determined with a Geiger-Müller counter, the sample being covered with just sufficient aluminum to absorb the Po^{210} alpha-particles. This residual activity showed one component of 6.4-day half-life (apparently Bi^{206} from the alpha-decay of At^{210}), and a second long-lived component (Bi^{207} from the alpha-decay of At^{211}).

Sufficient activity was available to determine absorption curves in aluminum and beryllium of both of these activities. The absorption properties of the 6.4-day activity were found to be similar to those reported[1] for Bi^{206}. The absorption curve for the long-lived Bi^{207} activity is shown in Fig. 1, resolved into its several components. Curve "O" is the unresolved aluminum absorption curve, while curve "A" is that of the electromagnetic radiation obtained by filtering out the electrons with 475 mg cm^{-2} of beryllium. Curve "A" was then resolved into curve "B," which includes the K x-rays and gamma-rays, and curve "C," the L x-rays. Correction was made in these curves for the absorption of the L x-rays by the beryllium. Finally, curve "D" represents the electrons.

On other samples of Bi^{207} prepared in another manner beta-ray spectrometer measurements were made and further detailed discussion of the decay characteristics

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† This work was performed under the auspices of the AEC.
[1] Templeton, Howland, and Perlman, Phys. Rev. **72**, 766 (1947).
[2] H. M. Neumann and I. Perlman, Phys. Rev. **78**, 191 (1950).
[3] D. G. Karraker and D. H. Templeton, Phys. Rev. **81**, 510 (1951).
[4] E. L. Kelly and E. Segrè, Phys. Rev. **75**, 999 (1949).
[5] Sunyar, Alburger, Friedlander, Goldhaber, and Scharff-Goldhaber, Phys. Rev. **78**, 326 (1950); **79**, 181 (1950); D. E. Alburger, Brookhaven National Laboratory Report BNL-64 (July, 1950).
[6] Corson, Mackenzie, and Segrè, Phys. Rev. **58**, 672 (1940).

of Bi^{207} will be referred to the following section, where these data are treated.

The two remaining astatine samples were dissolved after the initial volatilization separation, and further purification was effected consisting of carrying on tellurium during reduction with SO_2, and extractions into isopropyl ether. An aliquot was then counted to determine the amount of astatine present. When the decay of astatine was complete, bismuth carrier was added, bismuth and polonium fractions isolated, and the bismuth fraction carefully purified to eliminate the possibility of contamination by other elements. The activity of the bismuth fraction was followed with a Geiger counter and showed both the 6.4-day and long-lived components, each with the characteristics as noted in the chemically unseparated decay products of the astatine.

The half-life of Bi^{207} may be determined by measuring the activity which results from the decay of a known quantity of its parent, At^{211}. However, one encounters here the usual difficulty of estimating the counting efficiency for nuclei which decay by electron capture. In estimating the counting efficiency of Bi^{207} we have assumed that each electron capture decay event produces one L x-ray and that in addition each conversion electron is accompanied by an L x-ray. The Auger coefficient for L x-rays in this region is taken to be 0.5 and the counting efficiency of the x-rays has been shown to be about 3 percent in the counters employed. With these assumptions and estimations, the actual disintegration rate can be determined; and, therefore, the half-life of Bi^{207} can be calculated from the activities of At^{211} and the Bi^{207} daughter. The value so obtained is 50 years.

The counting rate of Bi^{207} without absorbers is largely that of the hard electrons. From the curves of Fig. 1 it can be shown that hard electrons are found in about 15 percent of the disintegrations and that the over-all counting efficiency without absorbers is about 1 count for 6 disintegrations.

Another sample of Bi^{207} prepared in a different manner was followed for decay over a period of 33 months. The minimum half-life, estimated from the counting data which indicated no significant decay, was 40 years. This means that the counting efficiency for Bi^{207} cannot be much lower than that estimated (15 percent) which corresponds with a 50-year half-life since the half-life decreases with the counting efficiency.

III. Bi^{207} FROM DEUTERONS ON LEAD

Bombardment of a thick lead target with 18-Mev deuterons should produce in various amounts all of the bismuth isotopes in the mass range 203 to 209. With the exception of Bi^{207} and Bi^{208}, all of these isotopes had been identified previously, and the longest-lived of these[3] is 14.5-day Bi^{205}.

In August, 1946, a thick lead target was bombarded with 18-Mev deuterons, and in November, 1947, the remaining bismuth activity was separated by J. J. Howland of this laboratory. The sample has shown no decay in the past 33 months, and from the counting statistics we could set a lower limit of 40 years for the half-life as mentioned above.

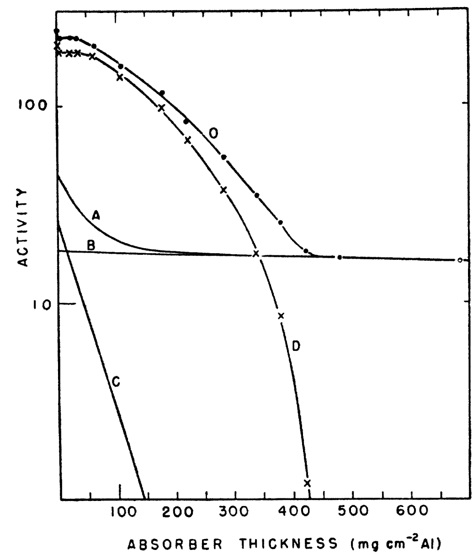


FIG. 1. Aluminum absorption curves for Bi^{207}. The components are: O, the unresolved curve; A, x-rays and γ-rays; B, K x-rays and γ-rays; C, L x-rays; D, electrons. See text for method of resolution.

The aluminum absorption characteristics for this activity were identical to those of Bi^{207} shown in Fig. 1. It is also significant that the ratio of electrons to L x-rays is the same in both samples, since if Bi^{208} contributed to the activity, an increased number of x-rays would be expected over those present in the pure Bi^{207} sample. (See discussion below on properties of Bi^{208}.) The long-lived activity formed in deuteron bombardment of lead is then essentially all Bi^{207}.

Sufficient activity from the deuteron bombardment of lead was available for use in a beta-ray spectrometer, and twelve conversion lines were observed. It is assumed that all of these lines belong to Bi^{207}; further arguments against attributing any of the electron radioactivity to Bi^{208} will be given below. The beta-ray spectrometer was a 255°-shaped magnetic field instrument operated with a resolution of 1.5 percent. The conversion lines, their assignments and relative intensities are shown in Table I. The absorption curve of Fig. 1 is consistent with these data, since it shows a major electron component at about 1 Mev and the tail of a soft component. The conversion lines around 500 kev are not sufficiently abundant to show up in the curve, and the hardest electrons are also in low abundance and blend with the electromagnetic radiation. The last four electrons listed in Table I were in too low abundance for us to have observed the L conversion lines.

There is no unique decay scheme which can be deduced from these data. It would be helpful to have the photo-electron spectrum from the uncoverted gamma-rays and their abundances, but not nearly

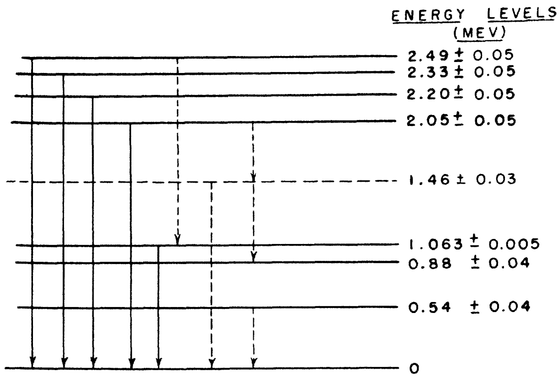


FIG. 2. Energy levels of Pb^{207}. Deduced from conversion electron spectrum of Bi^{207} and alpha-particle spectrum of Po^{211}.

enough activity could be made for such measurements. It may be of value, nevertheless, to indicate some possible arrangements of the energy levels of Pb^{207}, particularly since some independent data are available to aid in selections; and this has been done in Fig. 2. There is, of course, no way of deducing from the conversion electron spectrum the total electron capture decay energy even if the excited states of Pb^{207} resulting from the electron capture are as shown in Fig. 2. As a result, we do not know how much the decay energy exceeds 2.49 Mev, the energy of the hardest gamma-ray. The reasons for the energy level scheme as shown in Fig. 2 will be given after some further data are discussed which were obtained from the alpha-decay of Po^{211}.

It is readily apparent that the electron capture decay of Bi^{207} is highly forbidden. This is not surprising, as the spin of Pb^{207} is known to be $\frac{1}{2}$, and the spin of Bi^{207} would be expected to be the same as that of Bi^{209}, 9/2.

IV. ALPHA-DECAY OF Po^{211}

While about 40 percent of the At^{211} disintegrations are alpha-decay processes going to Bi^{207}, the remainder form Po^{211} by electron capture.[6] Being short-lived (estimated half-life several milliseconds), the Po^{211} exists in equilibrium with its parent and decays with the half-life of the latter. Some At^{211}, free from At^{210}, was

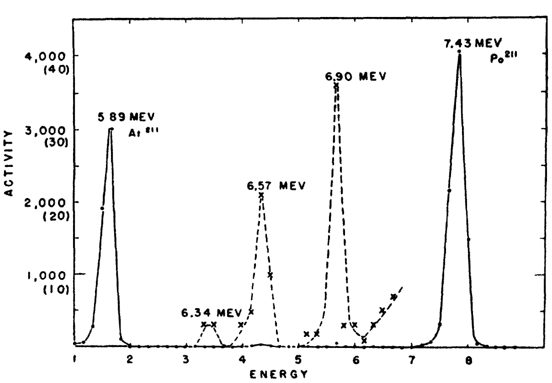


FIG. 3. Alpha-particle pulse analysis of At^{211}-Po^{211}. Broken-line portion of curve is on 100-fold expanded ordinate scale.

prepared by bombardment of a thin bismuth target with 25-Mev helium ions. A sample was collected by volatilization onto a platinum disk and examined with an alpha-particle pulse analyzer.[7] In addition to the alpha-particles of 5.89 Mev from At^{211} and 7.43 Mev from Po^{211}, three additional groups of low abundance were observed. These results are shown in Fig. 3, in which the groups in question are shown with a 100-fold expanded ordinate scale over that for the main groups. The low abundance groups are believed to be short range groups from Po^{211} for which the observed abundances and energies are given in Table II. The alpha-energies of At^{211} and of the Po^{211} ground-state transition served as standards to determine the energies of the short-range groups.

The branching of At^{211} decay is, from these experiments, 40.9±0.5 percent by alpha-emission and 59.1 ±0.5 percent by electron capture.

The principal piece of evidence that the three new alpha-groups are low energy groups of Po^{211} comes from their association with At^{211} and decay with the proper

TABLE I. Conversion electrons from Bi^{207}.

Electron energy (Mev)	Assignment	γ-ray energy (Mev)	Relative intensity of electron
0.049	*K* or *L*	0.137 or 0.064	0.11
0.062	Auger electrons		0.05
0.477	*K*	0.565	0.07
0.551	*L*		0.008
0.975	*K*	1.063	0.37
1.050	*L*		0.08
1.37	*K*	1.46	0.011
1.46	*L*		0.004
1.96	*K*	2.05	0.0014
2.11	*K*	2.20	0.0005
2.24	*K*	2.33	0.0014
2.40	*K*	2.49	0.0016

half-life. It is also of interest to examine whether or not the abundances of the several groups conform with what would be expected from alpha-decay theory. The test should not be expected to give close agreement, since it has been shown[8] that nuclei with odd nucleons will be characterized by prohibited alpha-decay which varies from one nucleus to another. Figure 4 shows the curve calculated[9] for the *half-life–decay energy* relationship for *even-even* polonium isotopes making the assumption of normal nuclear radii as previously[9] defined. Po^{211} would be expected to lie above the curve on two counts: (1) forbidden alpha-decay because of the odd nucleon, and (2) there is reason to believe that the nuclear radius is abnormally low. The half-life for Po^{211} has been estimated[10] to be 5 msec; but this is close to the half-life expected if there were no prohibition of alpha-decay,

[7] Ghiorso, Jaffey, Robinson, and Weissbourd, *The Transuranium Elements: Research Papers* (McGraw-Hill Book Company, Inc., New York, 1949), Paper No. 16.8, National Nuclear Energy Series, Plutonium Project Record, Vol. 14B.
[8] Perlman, Ghiorso, and Seaborg, Phys. Rev. **77**, 26 (1950).
[9] I. Perlman and T. J. Ypsilantis, Phys. Rev. **79**, 30 (1950).
[10] Curie, *et al.*, Revs. Modern Phys. **3**, 427 (1931).

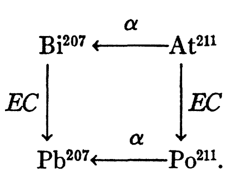
and we are inclined to estimate it as 50 msec or longer. From this the partial half-lives for the low energy groups are calculated from their abundances and plotted in Fig. 4. It may be seen that the relationships for these groups are not out of line with their assignment to short range groups of Po^{211}.

The energies of these alpha-groups of Po^{211} have an obvious bearing on the energy levels of Pb^{207}.

V. ENERGY LEVELS OF Pb^{207}

The three low energy alpha-groups from Po^{211} establish three of the energy levels of the daughter nucleus Pb^{207}. After the measured particle energies were converted into decay energies by adding the daughter nucleus recoil energy, differences from the ground-state transition gave the following levels for Pb^{207}: 0.54 $\pm$0.04, 0.88$\pm$0.04, and 1.11$\pm$0.06 Mev. In Table I, which shows the conversion electron lines from the decay of Bi^{207}, two of the gamma-rays can be seen to agree with two of these levels; but there are no conversion lines which correspond directly to the 0.88-Mev level. It is, of course, possible that those events which reach the 0.88-Mev level are either rare or that the degree of internal conversion of the gamma-ray is too low for detection. That this state does exist is known from the beta-decay of AcC″ (Tl^{207}) in which a gamma-ray of 870 kev in low abundance has been reported.[11]

Other levels and transitions indicated in Fig. 2 were derived from the conversion electrons of the Bi^{207} decay. To aid in the construction of the energy level diagram it is advantageous to estimate the decay energy of Bi^{207}. The basis of the estimation is the following decay cycle:



The alpha-decay energies of At^{211} and Po^{211} are known to be 6.0 Mev and 7.6 Mev, respectively. We shall estimate the electron capture decay energy of At^{211} using the empirical correlations of Thompson,[12] which gives 0.8 Mev as the decay energy corresponding to the measured half-life. From these values, the decay energy of Bi^{207} may be seen to be 2.4 Mev. Great reliance cannot be placed in this estimation, and it will be noted (Fig. 2) that a gamma-ray of greater energy (2.49 Mev) has been observed. However, the estimation is probably good enough to show that the hard gamma-rays cannot be in cascade and, therefore, they must lie in some such fashion as is shown in Fig. 2.

The broken lines in the diagram indicate alternative levels and transitions. For example, it is not necessary to assume a level at 1.46 Mev, since the same gamma-ray energy can be obtained between two other known levels. Similarly, the gamma-ray of 0.565 Mev can result from at least three transitions; but a state about this far above the ground state must exist, according to the alpha-decay data. The soft gamma-rays have not been placed in the diagram, because the energies are not known unambiguously. It is probable that the lowest excited state is that obtained from alpha-decay at 0.54 Mev; therefore, soft gamma-rays represent transitions between excited states. If there were any low lying excited states, it is probable that the corresponding alpha-groups would have been seen.

The apparent absence of low lying energy states of lead isotopes is explained by the assumption of a "closed shell" of 82 protons; and there is also, no doubt, a contribution from the neutron shell in this region closed with 126 neutrons. The lowest excited state of Pb^{204} is probably 374 kev above the ground state with a metastable state 905 kev above it.[5] Measurements of alpha-radioactivity of Po^{210} would indicate that the lowest lying excited state of Pb^{206} is 800 kev above the ground state, and this receives some support by the absence of gamma-radiation in the β^- decay of Tl^{206}. Of the other stable lead isotopes, the levels of Pb^{207} are discussed in this paper, while Pb^{208} is thought to have its lowest excited state at 2.62 Mev.

TABLE II. Abundances of alpha-particle groups from Po^{211}.

Alpha-particle energy (Mev)	Abundance (percent)
7.43	98.88
6.90$\pm$0.04	0.57
6.57$\pm$0.04	0.48
6.34$\pm$0.06	0.07

VI. PREDICTED PROPERTIES OF Bi^{208}

In the preparation of Bi^{207} with deuterons on lead it would be expected that Bi^{208} would be formed in comparable quantities. Nevertheless, we have attributed all of the electrons observed in the preparation to Bi^{207} as a result of considerations already given and some which follow.

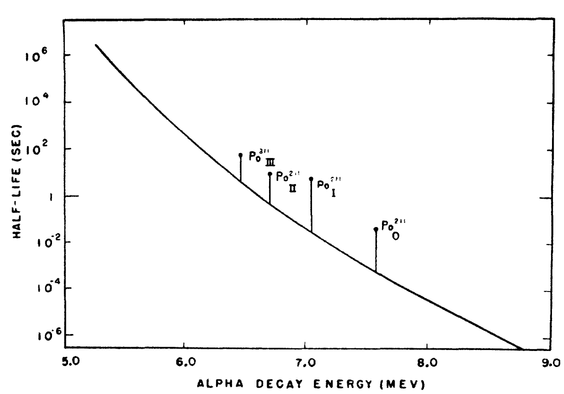
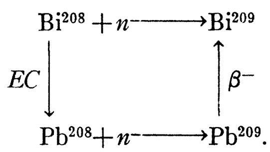


FIG. 4. Alpha-energy–half-life relationship for Po^{211} groups. Solid line is theoretical curve for even-even polonium nuclei on the assumption of normal nuclear radii.

[11] J. Surugue, J. phys. radium 7, 145 (1946).
[12] S. G. Thompson, Phys. Rev. 76, 319 (1949).

There is no decay cycle comparable to that used for Bi^{207} which has enough known components to permit estimation of the decay energy of Bi^{208}. However, in this case, some neutron binding energies are known which permit the closing of another cycle:



The β^- decay energy of Pb^{209} is known to be 0.70 Mev, and the neutron binding energies in Bi^{209} and in Pb^{209} have been measured although there are some uncertainties in the values. These binding energies from several sources have been discussed[13] and the principal uncertainty pertinent to the present discussion is the choice for the binding energy of the $Pb^{208-209}$ neutron between the limits 3.87 Mev and approximately 400 kev higher. If the higher value is selected, the electron capture energy for Bi^{208} becomes 2.3 Mev, while if the lower value is accepted, the decay energy is 2.7 Mev. From detailed work on the gamma-ray spectra of the natural radioactivities it is known that the lowest excited state of Pb^{208} is 2.62 Mev. It is seen, therefore, that the electron capture decay of Bi^{208} either cannot go to an excited state of Pb^{208} or can do so with very low decay energy. A search for the 2.62-Mev gamma-ray in the Bi^{207}, Bi^{208} mixture did not yield any positive results. As a result, we are probably justified in considering that all electrons of energy above the Auger electron region belong to Bi^{207} and that the only electromagnetic radiation from the decay of Bi^{208} is x-rays. We have made the tacit assumption in this discussion that Bi^{208} is long-lived, but obviously there is no direct evidence on this point.

We wish to thank Mr. C. I. Browne and Mr. G. D. O'Kelley for their aid in determining the electron spectra reported here, and Mr. A. Ghiorso for the alpha-particle pulse analyses.

[13] Huizenga, Magnusson, Simpson, and Winslow, Phys. Rev. **79**, 908 (1950).

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On the Systematics of Even-Even Alpha-Emitters

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The systematics of even–even alpha-emitters are studied. Empirical relationships are found, of the form

$$\log\lambda = a + b/v, \quad Z \text{ constant},$$

for values of $Z=84$, 86, 88, 90, 92, and 94. For these cases, the relationships found represent a more precise empirical correlation of alpha-energy and disintegration constant than does the Geiger-Nuttall rule. Relationships of the form

$$\log\lambda = A + B/v, \quad (A-2Z) \text{ constant},$$

exist for members of different partial radioactive series and are related to the disintegration schemes of the thorium and uranium families.

Values of the nuclear radii and internal potentials are calculated from the rigorous form of the Gamow-Condon-Gurney theory. The radii of "normal" even-even alpha-emitters can be presented by

$$r_0 = 1.57A^{1/3}\times 10^{-13} \text{ cm},$$

while the internal potential, U, can be represented by

$$E-U=0.52 \text{ Mev}.$$

The latter relationship is an indication of the approximate constancy, for the different nuclei, of the logarithmic derivative of the radial wave function at the boundary of the nucleus.

The empirical relations $\log\lambda = a+b/v$, Z constant, are compared with theory; and the significance of the comparison is discussed.

The values of the half-lives of Th^{224}, Ra^{220}, and Em^{216} are predicted.

The isotopes of polonium and emanation, which show departures from "normal behavior," are considered.

I. INTRODUCTION

THE amount of available experimental information concerning alpha-radioactivity has been greatly increased during recent years.[1,2] It is possible, therefore, to examine critically existing empirical relationships, such as the Geiger-Nuttall rule, to look for additional relationships, and to study more closely the validity of the Gamow-Condon-Gurney theory of alpha-decay. Although several investigations of the systematics of alpha-radioactivity have been made,[2–7] the present treatment is based on a somewhat different approach from that of any of the others.

The present study is limited to the ground state transitions of even-even alpha-emitting nuclides in

* Work performed at the Brookhaven National Laboratory, under the auspices of the AEC.

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[5] S. Biswas, Phys. Rev. **75**, 530 (1949).

[6] S. Biswas, Indian J. Phys. **23**, 51 (1949).

[7] I. Perlman and T. J. Ypsilantis, Phys. Rev. **79**, 30 (1950).