

latter value is in good agreement with the empirical value while the estimate for $g_L = Z/A$ is too small by a factor three. Although the approximations involved in this estimate are somewhat crude, it seems difficult to obtain a sufficiently large value of Δ for $g_L = Z/A$, even under rather extreme assumptions regarding the values of $(\kappa_s)_{Av}$ and $(\kappa_L)_{Av}$ in the two isotopes.

In this connection it may be mentioned that the Schmidt model is also favored by other evidence regarding nuclear magnetic moments. In fact, as is well known, the g_I -values of all odd nuclei fall within the limits given by the Schmidt model, whereas the g_I -values of a number of nuclei with high spin fall outside the limits predicted by the model in which $g_L = Z/A$. Moreover, reference may be made to the recent successes achieved by the individual particle model of nuclear structure in accounting for the angular momenta of nuclei.

Measurements of the h.f.s. anomalies in other elements would be desirable. There exist a number of odd elements (e.g., Sb, Eu, Ir) having isotopes with widely different g_I -values and for which the value of Δ according to (22) should be appreciable. In the case of even elements with isotopes of odd atomic number (22) gives a vanishing Δ for the Schmidt model, whereas for $g_L = Z/A$, values of Δ of more than one percent would be expected in several cases (e.g., Yb and Hg).

A number of elements (e.g., Na and K) have isotopes of odd numbers of protons and neutrons whose spin and h.f.s. have been measured. These nuclei are of special interest for the problem of nuclear structure, and a study of their h.f.s. anomalies might give some indication regarding the composition of their moments.

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On the α - β - and α - K -Branching of the Heaviest Natural and Artificial Radioactive Substances*

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Based on simple empirical regularities in α -decay properties of the heavy elements, the following substances which show β -emission or K -capture should also be α -radioactive with α - β - or α - K -branching ratios of at least 10^{-10} : MsTh_2 , MsTh_1 , AcK ; $_{88}\text{Ra}^{228}$, $_{91}\text{Pa}^{233}$, $_{93}\text{Np}^{234}$, $_{93}\text{Np}^{238}$, $_{93}\text{Np}^{236}$, $_{93}\text{Np}^{238}$, $_{93}\text{Np}^{239}$, $_{94}\text{Pu}^{241}$, $_{95}\text{Am}^{242}$.

I. INTRODUCTION

IN addition to the long known dual disintegration of the C-bodies of the three natural radioactive disintegration series, the existence of a number of other double disintegrations has been established. M. Perey¹ succeeded in proving that actinium emits a weak α -radiation. This disintegration creates element 87 (francium). B. Karlik's and T. Bernert's² proof of the dual disintegration of RaA made it possible to establish the occurrence in the natural disintegration series of element 85 (astatium) through the isotope $_{85}\text{At}^{218}$.** Prior to this, element 85 has already been artificially obtained and chemically investigated through reaction $_{83}\text{Bi}^{209}(\alpha, 2n)_{85}\text{At}^{211}$ by D. R. Corson, K. R. McKenzie, and E. Segre.³ This element shows α - K -branching. In

the newly established $4n+1$ -series $_{83}\text{Bi}^{213}$ suffers an α - β -decay.⁴

II. EMPIRICAL REGULARITIES OF THE α -DISINTEGRATION OF THE HEAVIEST NUCLEI

In the range of α -instability of the heaviest nuclei all elements emitting β -rays or showing K -capture must also be α -radioactive. Based on simple empirical regularities, therefore, branching ratios for some of these radioactive elements which have not as yet been measured are stated below. However, only dual disintegrations with branching-ratios of 10^{-10} or higher will be mentioned.

If the disintegration energies of the natural α -radioactive elements are plotted in a diagram as function of their mass numbers⁵ (Fig. 1), a family of curves is formed by lines connecting nuclei of the same atomic

* This paper was written in August, 1947.

¹ M. Perey, J. de phys. et rad. **10**, 435 (1939). M. Perey and Lecoq, J. de phys. et rad. **10**, 439 (1939).

² B. Karlik and T. Bernert, Zeits. f. Physik **123**, 51 (1944).

** In the case of ThA and AcA , also, long range α -particles had been observed.² The simplest explanation would be a dual decay of ThA and AcA . But applying energy-conservation rules it follows that there exist considerable difficulties in accepting this interpretation.

³ Corson, McKenzie, and Segre, Phys. Rev. **57**, 1087 (1940).

⁴ Hagemann, Katzin, Studier, Ghiorso, and Seaborg, Phys. Rev. **72**, 252 (1947); English, Cranshaw, Demers, Harvey, Hincks, Jelley, and May, Phys. Rev. **72**, 253 (1947).

⁵ J. Schintlmeister, Oesterreichische Chemiker Zeitung Nr. 17, 1938, Nr. 9/12, 1943. Regularities in Geiger Nuttall diagrams referring to the individual values of Z were pointed out by Berthelot in the J. de phys. et rad., serie VIII, **3**, 17 (1942) and by N. Feather, Proc. Roy. Soc. Edinburgh, **62**, 211 (1946).

number. Another family of curves occurs if nuclei with the same neutron-proton differences, $N-Z$, are connected with each other. Of course, other variables can be chosen as abscissae; for instance, the number of the neutrons N , the atomic number, Z , $N-Z$ or N/Z . In each case, similar families of smooth curves are again obtained, for appropriate parameters.

The atom nuclei may be separated into three types in accordance with the number of their constituent protons and neutrons: Even-even ($e-e$)-types (both particle numbers even), even-odd ($e-o$)-types (one particle number odd) and odd-odd ($o-o$)-types (both particle numbers odd). From Fig. 1 can be seen that the ($e-e$)-nuclei (marked $\bullet\bullet\bullet$) lie directly on the curves, while the ($e-o$)-nuclei (marked $\times\times\times$) and the ($o-o$)-nuclei (marked $\circ\circ\circ$) are almost invariably situated slightly below or above the curves.

By means of this diagram, the energy of α -rays can be predicted with a fair degree of accuracy for the even-even nuclei and with a little less accuracy for the ($e-o$)- and ($o-o$)-nuclei.

One would like to establish with the greatest possible accuracy the corresponding half-life periods, since they must be exactly known in order to ascertain the value of the branching ratio. It is not advisable to determine, with the aid of the Geiger-Nuttall principle, half-lives from the known α -ray energies since this law is of only approximate validity. A calculation with the aid of the Gamow formula does not show very accurate results

either, because some of the parameters which enter into the formula vary from nucleus to nucleus in a manner as yet unknown.

For this reason, a second diagram has been inserted into Fig. 2, which again shows as abscissa the mass number. However, the common logarithm of the half-life is used as ordinate, in place of the disintegration energy. The regularities thus obtained resemble those in Fig. 1. All ($e-e$)-nuclei lie on smooth curves, joining the nuclei with the same atomic number or with the same number of excess neutrons. Generally the ($e-o$)- and ($o-o$)-nuclei again lie off these curves. They are almost invariably situated below these curves. But, as Fig. 2 shows, the ($e-o$)- and ($o-o$)-nuclei lie with a fair degree of accuracy on separate smooth curves, which are marked by dotted lines. For the radioactive bodies with atomic numbers 83, 84, 85 the differences between ($e-e$)- and ($e-o$)- or ($o-o$)-nuclei seem to be very small.

The artificially produced α -radioactivities such as $^{211}_{85}\text{At}$, the members of the $4n+1$ -series⁴ and the α -radioactive elements⁶ with atomic numbers larger than 92 (according to data published to date) are easily classified in Figs. 1 and 2.

III. THE DUAL DISINTEGRATIONS OF THE NATURAL RADIOACTIVE ELEMENTS: MsTh_2 , MsTh_1 , $^{223}_{87}\text{Fr}$ (AcK) AND RaE

Of the natural elements emitting α -rays, only MsTh_2 , MsTh_1 , and AcK show dual disintegration probabilities

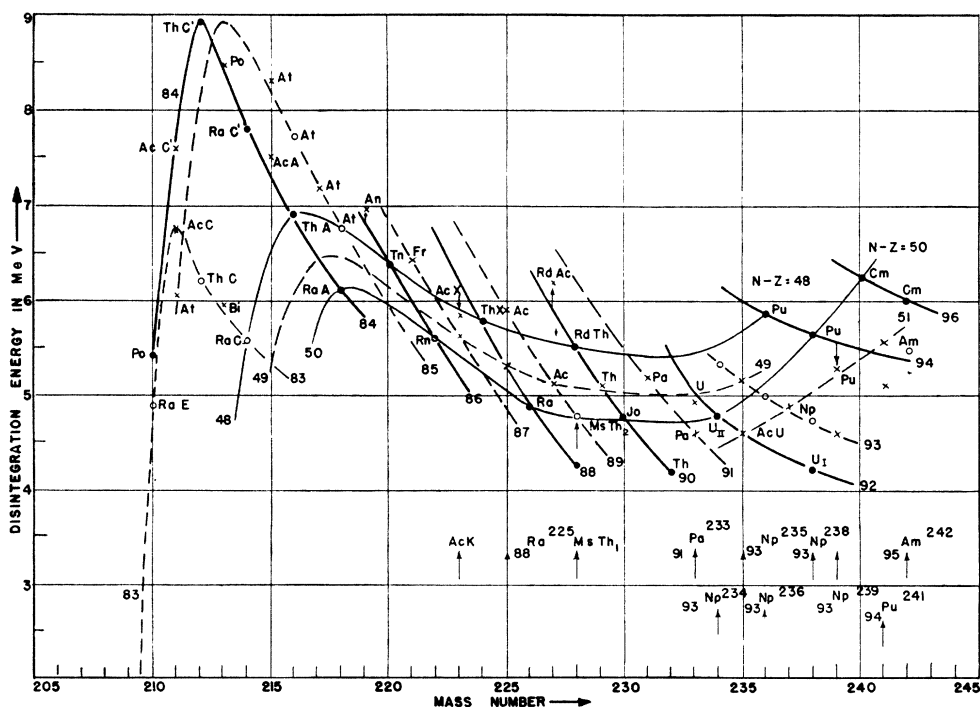


FIG. 1. α -Radioactive elements: disintegration energy and mass number. Even-even nuclei $\bullet\bullet\bullet$, even-odd nuclei $\times\times\times$, odd-odd nuclei $\circ\circ\circ$.

⁶ Seaborg, James, Florin, Hopkins, Studier, Hyde and Ghiorso, Science, **104**, 379 (1946); Seaborg, James, and Morgan, Chem. Eng. News **25**, 358 (1947); James M. Cork, Radioactivity and Nuclear Physics (New York, 1947).

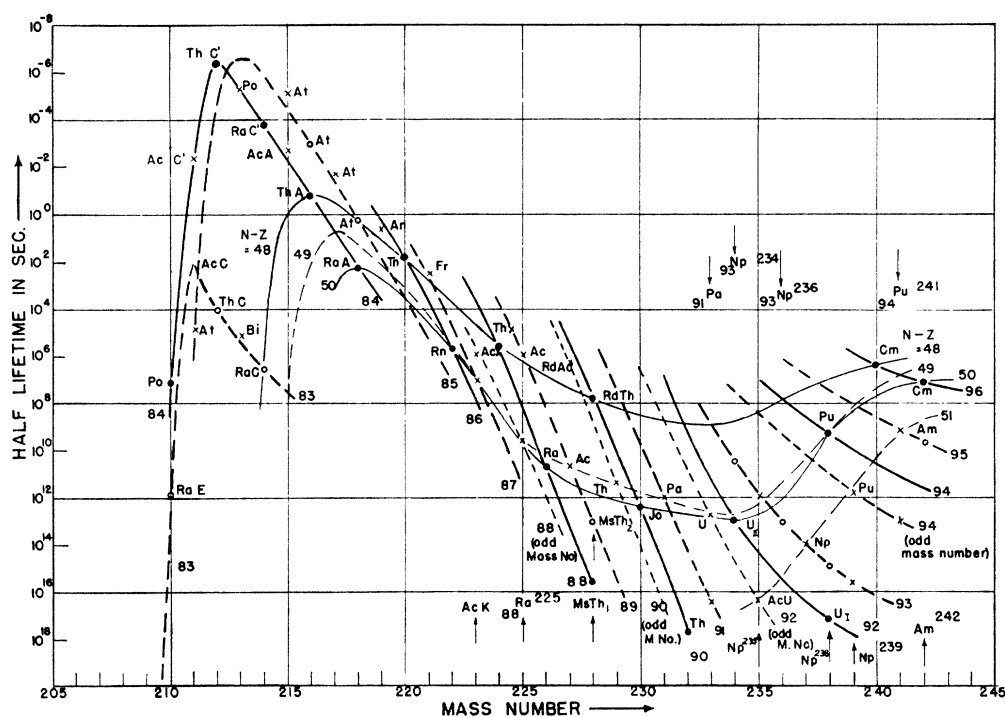


FIG. 2. α -radioactive elements: half-life and mass number. Even-even nuclei $\bullet\bullet\bullet$, even-odd nuclei $\times\times\times$, odd-odd nuclei $\circ\circ\circ$.

as yet not observed whose branching ratios are higher than 10^{-10} . Recently, the dual disintegration of radium *E* was experimentally demonstrated, as can be seen from a very exhaustive investigation which was published while this paper was being prepared.⁷ Data concerning the mentioned radioactive elements have been compiled in Table I. Column 1 indicates the element in question, column 2 the half-life of the β - or *K*-radiation in seconds, column 3 the estimated limits of the half-life of the supposed α -activity, columns 4 the estimated

limits of the α - β -branching ratio, column 5 the most probable value of the α -energy.

The limits of accuracy were estimated according to the following points of view:

As far as even-even nuclei are concerned, the values of the disintegration energy and of the half-life are uncertain only if it is necessary to extrapolate the curves into regions where no measured values are as yet available. This is the case with MsTh_1 . In spite of this, however, the branching ratio can be stated with an accuracy of, perhaps, a factor of 10.

Generally, as can be seen from the diagram, the uncertainty of the α -decay energy amounts to 0.2 Mev while the uncertainty of the half-life amounts to a factor of ten.

Experimentally determined values are, of course, given for RaE in Table I.

Disintegration Schemes and Experimental Possibilities of Proof

Attempts to demonstrate the dual disintegration of MsTh_2 have already repeatedly been made. The disintegration scheme reads as follows:

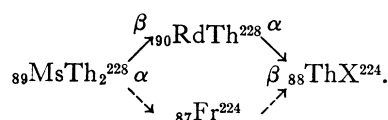


TABLE I. Branching ratios and α -decay energies of some natural and artificial radioactive substances.

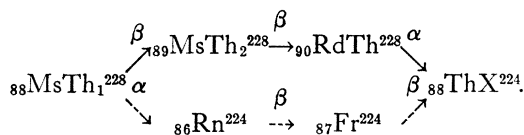
Element	Half-life of β - or <i>K</i> -radiation in sec.	Lower and upper limit of the half-life of the supposed α -radiation in sec. values from Fig. 2	Lower and upper limit of the α - β - or α - <i>K</i> -branching ratios	α -disintegration energy in Mev. values from Fig. 1
Natural-radioactive substances				
MsTh_2	$\beta^- 2.026 \times 10^4$	$3 \times 10^{12} - 3 \times 10^{13}$	$6 \times 10^{-9} - 6 \times 10^{-10}$	4.75 ± 0.1
MsTh_1	$\beta^- 2.11 \times 10^8$	$2 \times 10^{15} - 2 \times 10^{16}$	$10^{-7} - 10^{-8}$	4.25 ± 0.1
AcK	$\beta^- 1.26 \times 10^3$	$2 \times 10^6 - 3 \times 10^7$	$6 \times 10^{-4} - 4 \times 10^{-5}$	5.5 ± 0.2
RaE	$\beta^- 4.32 \times 10^4$	4×10^9	5×10^{-7}	4.87
Artificial-radioactive substances				
$^{88}\text{Ra}^{226}$	$\beta^- 1.21 \times 10^8$	$10^9 - 2 \times 10^{10}$	$10^{-3} - 6 \times 10^{-5}$	5.2 ± 0.2
$^{91}\text{Pa}^{233}$	$\beta^- 2.367 \times 10^6$	$3 \times 10^{15} - 3 \times 10^{16}$	$8 \times 10^{-10} - 8 \times 10^{-11}$	4.50 ± 0.2
$^{92}\text{Np}^{235}$	$\beta^- 3.80 \times 10^5$	$\sim 10^{10}$		5.3
$^{92}\text{Np}^{235}$	$\beta^- 2.07 \times 10^7$	$\sim 5.10^{11}$		5.15
$^{92}\text{Np}^{235}$	$\beta^- 7.2 \times 10^4$	$\sim 5.10^{12}$		5.0
$^{92}\text{Np}^{235}$	$\beta^- 1.73 \times 10^5$	$\sim 10^{14}$		4.7
$^{92}\text{Np}^{239}$	$\beta^- 2.01 \times 10^5$	$\sim 10^{15}$		4.6
$^{94}\text{Pu}^{241}$	β^-	$\sim 10^{13}$		5.1
$^{96}\text{Am}^{242}$	$\beta^- 6.48 \times 10^4$	$\sim 10^{11}$		5.5

⁷ E. Broda and N. Feather. Proc. Roy. Soc. **190**, 20 (1947).

Of all the experiments conducted to date in an at-

tempt to show the dual decay of MsTh_2 ⁸ none has even approached the sensitivity of the method used by Hahn and Erbacher.^{9,10} By applying chemical separation methods, element 87 was isolated and examined for β -radiation. Negative results obtained places an upper limit on the α - β -branching ratio of MsTh_2 , dependent upon the assumed value of the half-life of element 87. In Table II are shown the results obtained by Hahn and Erbacher. It appears in comparison with the value of Table I that the sensitivity has been just not large enough for a positive confirmation of the α -radiation of MsTh_2 if the β -half-life of the element ${}_{87}\text{Fr}^{224}$ is longer than 10 minutes. Applying energy-conservation rules we obtain for the value of the β -disintegration energy of ${}_{87}\text{Fr}^{224}$ 3.2 Mev. By means of the Sargent diagram it is possible to estimate the half-life of ${}_{87}\text{Fr}^{224}$ to 50 seconds for an allowed transition, for a "first forbidden transition" to 1.4 hours.

The disintegration of MsTh_1 occurs as follows:



In spite of the higher α - β -branching ratio it seems that here, too, no direct confirmation of the α -radiation of MsTh_1 can be obtained because of the fact that it is impossible to prevent the presence of small traces of other α -rays with similar ranges. According to the above mentioned disintegration scheme the same isotope of element 87 is again being formed. In conformity with the estimates made here it can be concluded from these experiments by Hahn,¹⁰ which were conducted with a mesothorium-source, that the α - β -branching ratio of MsTh is lower than 10^{-4} . The β -half-life of ${}_{86}\text{Rn}^{224}$ can be estimated by means of energy considerations to 16 hours if we assume an allowed transition, to 60 days for a first forbidden transition.

A positive confirmation of the dual disintegration of MsTh_1 is perhaps possible if the very sensitive method used by Hahn and Erbacher is applied to very strong mesothorium-sources.

The disintegration scheme of $\text{AcK}({}_{87}\text{Fr}^{223})$ can be written as follows:

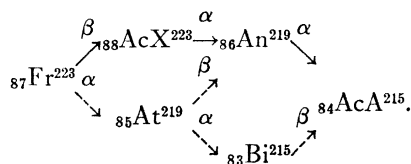
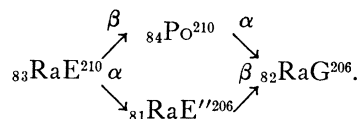


TABLE II. Results of Hahn and Erbacher for the maximal α - β -branching ratio α/β of MsTh_2 .

Supposed half-life of ${}_{87}\text{Fr}^{224}$	Maximal α - β -branching-ratio
10 minutes	2.9×10^{-8}
9.5 hours	5.5×10^{-9}
1 day	1.6×10^{-8}
10 days	5×10^{-9}
100 days	8×10^{-9}
10 years	1.5×10^{-7}

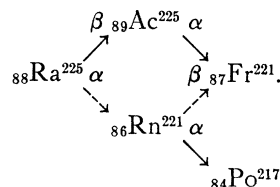
If it is possible to produce rather strong ${}_{87}\text{Fr}^{223}$ sources, the direct confirmation of the α -radiation of ${}_{87}\text{Fr}^{223}$ should not meet special difficulties. To complete the picture, the disintegration scheme of RaE , already experimentally confirmed, may be mentioned:



IV. THE BRANCHING RATIOS WITH ARTIFICIAL RADIOACTIVE SUBSTANCES

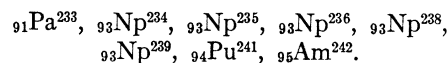
Concerning the newly established $4n+1$ series, ${}_{88}\text{Ra}^{225}$, in addition to ${}_{83}\text{Bi}^{213}$, should undergo a dual disintegration. Experimentally it has already been established that the α - β -branching ratio for ${}_{88}\text{Ra}^{225}$ is less than 1 percent. The values read off from Figs. 1 and 2 were also recorded in Table I. From this it can be seen that the branching ratio is lower than 10^{-3} .

The disintegration scheme reads:



Experimental confirmation should not meet any special difficulties.

In addition the following radioactive substances which show β -radioactivity or K -capture should be also α -radioactive with α - β - or α - K -branching ratios larger than 10^{-10} :



The values of the branching ratios for elements with atomic numbers larger than 82 are not entered in Table I, because the curves drawn in this range are very inaccurate due to the lack of published data.

Note added at proof:—While this paper was being prepared for publication, the following related articles appeared: Vigneron, *Comptes Rendus* **225**, 1067 (1947); Feather, *Reports Prof. Phys.* **11**, 19 (1948); Karlik, *Acta Phys. Austriaca* **2**, No. 2, 182 (1948); Perlman, Ghiorso, and Seaborg, *Phys. Rev.* **74**, 1730 (1948).

⁸ J. Schintlmeister and E. Rona, *Akad. Wiss. Wien* **147**, 49 (1938); see there also other literature.

⁹ O. Hahn and O. Erbacher, *Physik Zeits.* **27**, 531 (1926).

¹⁰ O. Hahn, *Naturwiss.* **14**, 159 (1926).