than the hard component; it is difficult to see how the shower correction might help to *raise* the ratio from 1.18 to 1.28 as would be expected assuming a lifetime of 2 μsec. The presence of lead between the counters as in Fig. 1 at the greater altitude does not alter this conclusion materially.

We shall now briefly consider our results in connection with the density effect of ionization losses recently discovered by Fermi.[9] This effect becomes important only at high energies. A mesotron having a residual range of 280 g/cm^2 of air (the path between Pian Rosà and Chatillon) has an energy of 5 or 6×10^8 ev. At these energies Fermi's effect is small and cannot account for the effects we have observed. Fermi himself states in his paper that the experiments with comparatively thin absorbers cannot be explained by the density effect alone. It seems therefore that the instability hypothesis is still necessary to explain these effects. A striking confirmation of this hypothesis is afforded by a photograph recently published by E. J. Williams.[15]

On the other hand it has been shown by Fermi that the existence of a density effect on ionization means that the value of the lifetime which Euler and Heisenberg have derived from the experiments of Ehmert (with large absorbing thicknesses) must be increased by about a factor 2. It is indeed remarkable that both our measurements and those of Pomerantz, which were made also with comparatively small absorbing thicknesses, indicate a rather large value of the proper lifetime of the mesotron. In another paper we shall describe evidence which we have collected on this problem by a different method, that is, by a detailed study of the soft component which accompanies the mesotrons near sea level. Here also we have not found indication of the presence of "disintegration electrons" in the number that would be expected if the lifetime of the mesotron were as low as 2 microseconds. This also is in partial agreement with experiments made on somewhat similar lines by Pomerantz,[13] who finds a value of 6 μsec. from the intensity of the "disintegration electrons."

In conclusion we wish to acknowledge our indebtedness to the "Comitato per la geofisica e meteorologia del Consiglio Nazionale delle Ricerche" for financial assistance.

[15] E. J. Williams and G. E. Roberts, Nature **145**, 102 (1940).

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The Missing Heavy Nuclei

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Considerations of the regularities in the distribution of isotopes lead to the following conclusions. (1) Rn, AcA, ThA, and RaA should all be somewhat beta-active but the branching ratios would be too small for detection of the activity except for Rn and RaA. (2) $_{93}$EkaRe^{239} should be beta-active with a roughly estimated half-life of about 1 month, $_{93}$EkaRe^{237} should be an alpha-emitting nucleus, and $_{92}$U^{237} should be beta-active. (3) The heaviest beta-stable isotopes of transuranic elements should be as follows: $_{93}$EkaRe^{237}, $_{94}$EkaOs^{244}, $_{95}$EkaIr^{243}, $_{96}$EkaPt^{250}. It is shown that isotopes of transuranic elements should undergo fission upon exposure to slow neutrons, which may account for their absence in nature. Their presumable greater probability for spontaneous fission might also account for their absence. The relative abundance of U^{235} and U^{238} is in fair agreement with the hypothesis that the amount of U^{235} was determined by a balance between production from U^{239} and loss by fission. The probable chain of disintegrations of $4n+1$ nuclei is discussed. The estimated half-lives of all are too short for them to have survived. Their absence is to be attributed to the absence of a possible long-lived transuranic ancestor. Either the hypothetical irradiation by neutrons or spontaneous fission of a transuranic ancestor will account for the low abundance of Bi^{209}.

In a letter to the editor[1] it has been pointed out that the absence in nature of atoms of atomic number greater than 92 can be accounted for on the basis of a hypothetical exposure of all matter to irradiation by neutrons in some early phase of

[1] L. A. Turner, Phys. Rev. **57**, 157 (1940).

the history of the universe. The absence of all radioactive atoms of mass $4n+1$ and of U^{236} would also be expected on this hypothesis. The purpose of this paper is to amplify these considerations and discuss other related conclusions concerning the radioactive properties of some of the heavy nuclei.

The explanation of nuclear fission first suggested by Meitner and Frisch[2] and later developed by Bohr and Wheeler[3] does not make it clear why the upper limit of the atomic number of elements occurring in nature should be 92. Although these heaviest nuclei approach the limit of stability for spherical nuclei at about $Z=100$, they still require about 5 Mev of additional energy for fission to occur without the necessity of penetration of a potential barrier by the heavy fragments. Atoms having Z's of 93, 94, 95, etc. would require less energy of excitation for instability, but this energy would nevertheless be considerable, so that there is no obvious reason why they should be entirely unstable. Uranium is, however, the only element occurring naturally which has an isotope which will undergo fission when it captures thermal neutrons. The theory of Bohr and Wheeler leads one to expect that even more of the isotopes of the transuranic elements would do this, so it is possible that this process may be responsible for their absence. In what follows it will be shown that all expectable atoms for $Z>92$ would undergo fission with reasonably slow neutrons (<0.5 Mev) if not with thermal ones.

Before entering upon the special discussion a few general points should be recalled. As is now well known the natural radioactive substances which emit beta-particles differ in no special way from the artificially produced beta-active substances except in the manner of their formation. The necessary conditions for occurrence or nonoccurrence of beta-activity are apparently the same throughout the periodic table. The emission of alpha-particles is the property of the naturally radioactive atoms which distinguishes them from the ordinary ones. This emission is possible because of a somewhat faster increase of mass with atomic number among the heavier atoms so that the removal of an alpha-particle is exothermic, rather than endothermic as for the lighter nuclei. It is apparently not an expression of any special new property of the nucleus considered by itself. The alpha-emitting nuclei are, therefore, the analogs of the stable lighter atoms as has been emphasized by Gamow and others. In what follows, such nuclei will sometimes be referred to as the beta-stable ones when the emphasis is being put upon this similarity to the stable lighter atoms. Since the conditions for emission of alpha-particles and of beta-particles are independent of each other it is not surprising that they are often both satisfied for the same nucleus and lead to branching, as with the C products. The necessary condition for the observation of such branching is that the two probabilities for disintegration be of nearly the same order of magnitude so that an appreciable fraction of the nuclei will disintegrate in the less probable way. It is probable that others of the beta-active atoms are also slightly alpha-active as actinium has been found to be[4] but that the alpha-activity is of even less relative importance because of the greater probability of beta-disintegration of most of them. The same consideration applies the other way around to the short-lived alpha-active nuclei. Some of them may have small probabilities for ejection of beta-particles which by themselves would give much greater half-lives, so that only a negligible fraction of the atoms do emit beta-particles. Several possible instances are discussed below. The important general point is that the existence of an alpha-activity of short half-life without observed beta-activity gives no assurance that the nucleus in question is beta-stable. The erroneous contrary assumption has sometimes been made in discussing the nuclei.

Further, it should be noticed that in all cases when a nucleus emits beta-particles the nucleus of the same Z of mass number greater by 2 units is also beta-active, and usually with a shorter half-life. The half-lives for emission of alpha-particles go the other way. The probability of ejection of an alpha-particle is less for the heavier of two nuclei of the same Z differing by 2 mass units with the exception of the three such

[2] L. Meitner and R. Frisch, Nature **143**, 239 (1939).
[3] N. Bohr and J. A. Wheeler, Phys. Rev. **56**, 426 and 1065 (1939).
[4] M. Perey, J. de phys. et rad. **10**, 435 (1939).

pairs which involve the lowest mass numbers of the heavy radioactive elements. These pairs are $_{84}Po^{210}$ and $_{84}Po^{212}$(ThC′), $_{83}Bi^{210}$(RaE) and $_{83}Bi^{212}$(ThC), $_{83}Bi^{209}$ and $_{83}Bi^{211}$(AcC).

Beta-Stable Nuclei of Even Elements

As a preliminary to the discussion of fission of the transuranic atoms it is desirable to see which mass-numbers of such nuclei would be expected to be beta-stable. The only basis for such considerations is the assumption that the general shape of the nuclear energy surface in the transuranic region can be obtained by extrapolation from that for the known nuclei. This means that the relations between the masses of the beta-stable atoms of $Z<92$ can be extended into the region for $Z>92$.

From the table of stable isotopes given by Livingood and Seaborg[5] have been taken the masses of the heaviest isotope for the even elements. These have been plotted in Fig. 1, with the addition of the masses of the heaviest beta-stable isotopes of Ra, Th and U. The quantity, $y=A+50-3Z$, has been plotted as ordinate. This quantity has no particular theoretical significance but is convenient because of the empirical fact that the mass of the heaviest isotope increases by nearly 3 units for an increase of unity in the atomic number, for the heavy atoms. From Fig. 1 it is apparent that the interpolated values of 214 and 220 are extremely probable ones for the masses of the heaviest beta-stable isotopes of $_{84}$Po and $_{86}$Em, respectively. Similarly, the two extrapolated points for $_{94}$EkaOs and $_{96}$EkaPt give 244 and 250 as highly probable upper limits for those elements. The true points might, of course, be lower to correspond to 244 and 248 or 242 and 248. The points for these nuclei and for all of the other known nuclei at the end of the periodic table have been plotted on a larger scale, but in the same way, in Fig. 2. The points for those heaviest beta-stable even nuclei lie along the line for $y=12$.

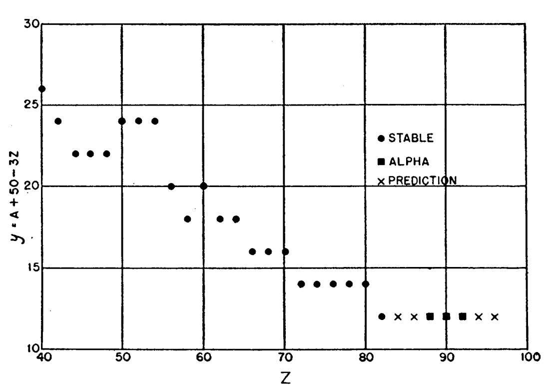

Fig. 1. Heaviest isotopes of even elements.

If this is the upper limit for beta-stability it means that Rn($_{86}Em^{222}$), AcA($_{84}Po^{215}$), ThA($_{84}Po^{216}$) and RaA($_{84}Po^{218}$) must all be somewhat beta-active. There are three known beta-active nuclei which lie along the line for $y=14$ with Rn and ThA. These, with their half-lives, are RaD($_{82}Pb^{210}$), ∼25 yr; Ms_1($_{88}Ra^{228}$), 5.7 yr; and UX_1($_{90}Th^{234}$), 24.5 days. If we assume a similar type of beta-disintegration (Sargent curve) we may estimate the beta half-lives of ThA and Rn by logarithmic interpolation to get 13 yr. and 7 yr., respectively. These correspond to branching ratios of 10^{-3} for Rn and 10^{-9} for ThA. The immediate descendant from Rn by beta-disintegration would be $_{87}EkaCs^{222}$. It lies on the $y=11$ line with beta-active $_{83}Bi^{210}$(RaE, 5 days), $_{89}Ac^{228}$(Ms_2, 6.13 hr.) and $_{91}Pa^{234}$(UX_2, 1.14 min.) and would thus be expected to be beta-active with a half-life of some hours. From curves given by Heisenberg[6] one can estimate the energy which would be released in alpha-disintegration of $_{87}EkaCs^{222}$ to be $\sim 9.4\times 10^{-6}$ erg. By the Geiger-Nuttall relation this leads to an estimated half-life of ∼12 min. so that by far the greater number of nuclei would be expected to emit alpha-particles to become $_{85}EkaI^{218}$ (not shown on Fig. 2. This paragraph has been revised in proof. The less probable chain of descendants from $_{87}EkaCs^{222}$ is indicated on Fig. 2, because the preponderance of its alpha-activity was originally overlooked.) $_{85}EkaI^{218}$ would be expected, by a similar argument, to be unstable with respect to emission of both beta- and alpha-particles, the latter predominating to give $_{83}RaC^{214}$ and with a half-life of ∼0.1 sec. The suggested beta-disintegration of RaA would also lead to $_{85}EkaI^{218}$. RaA lies on the $y=16$ line and is to be compared with $_{82}Pb^{212}$(ThB) which is beta-active with a

[5] J. J. Livingood and G. T. Seaborg, Rev. Mod. Phys. **12**, 30 (1940).

[6] W. Heisenberg, Solvay Congress, 1934, pp. 289 to 344, particularly Fig. 10 on p. 320.

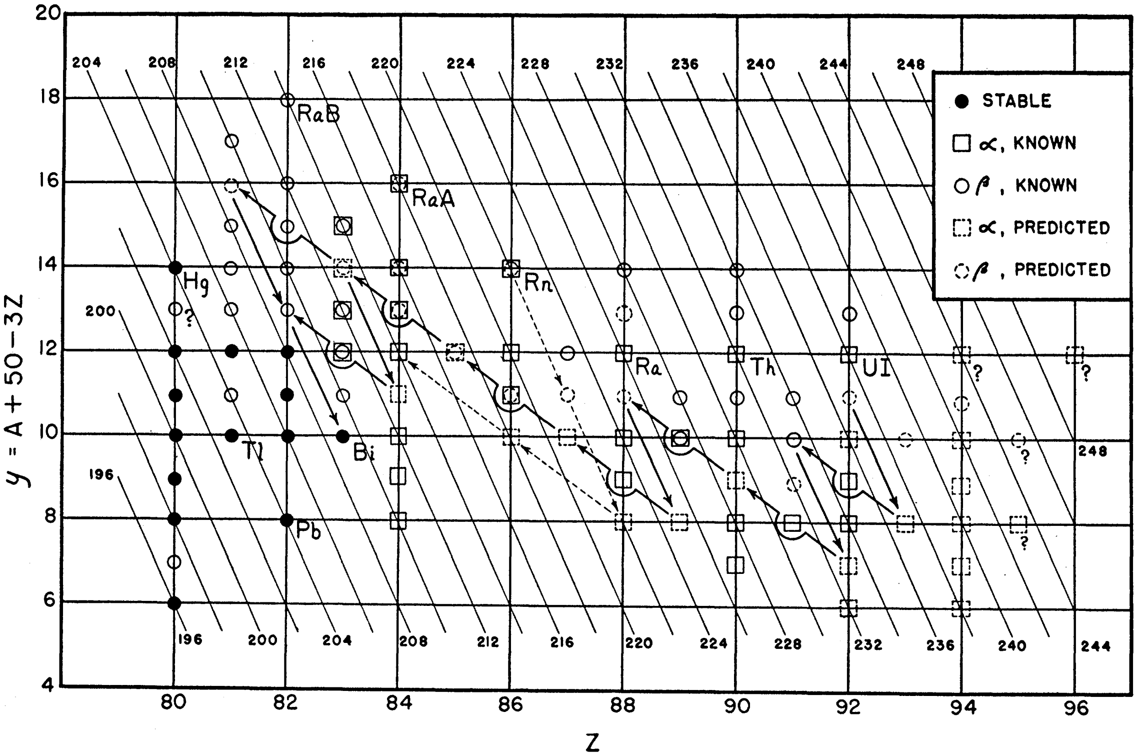

FIG. 2. Isotopes of the elements of high atomic number.

half-life of 10.6 hr. The probability of beta-disintegration of RaA presumably corresponds to a similar but somewhat shorter half-life, whereas the actual half-life determined by the alpha-activity is 3.05 minutes. This indicates a branching ratio somewhat greater than 5×10^{-3}. It is of interest in this connection that Hulubei and Cauchois[6a] have reported three x-ray lines of element 85 coming from the disintegration products of Rn. It is hard to see, however, how these lines could have been observed if the branching ratios of Rn and RaA are as small as here estimated. The detection of either the beta-particles or fast alpha-particles arising in such branching presents obvious difficulties because of their small number. The branching ratio for AcA, like that for ThA, would be negligibly small.

BETA-STABLE NUCLEI OF ODD ELEMENTS

The possible mass numbers of the beta-stable nuclei of the odd elements can be found by the following rule: The possible mass-numbers for the beta-stable isotopes of an odd element are $A+1$, $A-1$, and $A-3$, A being the heaviest beta-stable isotope of the preceding element. There is no exception to the rule among the stable elements from $_8$O to $_{83}$Bi inclusive, or among the known beta-stable radioactive elements.

This rule gives 227, 225, 223 as possible mass numbers for $_{89}$Ac. 227 is the mass number of ordinary beta-active Ac, 223 is ruled out because of the existence of beta-stable AcX($_{88}$Ra^{223}), assuming the improbability of occurrence of isobars of adjacent elements. 225, one of the $4n+1$ family, is left as the only probable beta-stable $_{89}$Ac.

The rule gives 233, 231, and 229 for $_{91}$Pa. The mass number of existing alpha-active Pa is 231. Pa^{229} is improbable because of the probable beta-stability of $_{90}$Th^{229}. This latter nucleus lies on the $y=9$ line between beta-stable AcX($_{88}$Ra^{223}) and AcU($_{92}$U^{235}). It is another of the missing $4n+1$ nuclei. Pa^{233} is the product of the dis-

[6a] H. Hulubei and Y. Cauchois, Comptes rendus **209**, 39 (1939).

integration of 25-min. beta-active Th^{233}, which is produced by the capture of slow neutrons by Th^{232}. Meitner, Hahn and Strassmann[7] found an active Pa having a half-life of 25 days, and attributed it to Pa^{233}. Additional support for the correctness of this conclusion is obtained as follows. This nucleus lies on the $y=10$ line of Fig. 2. The other known odd nuclei on the $y=10$ line near to it are ${}_{89}Ac^{227}$ and ${}_{93}EkaRe^{239}$ (see below). Ac^{227} is beta-active with a half-life of 13.5 yr. The ratio of this half-life to 25 days is 197. This agrees in order of magnitude with the ratios of two pairs of adjacent beta-active nuclei. These are: $Ms_2(Ac^{228})$ and $UX_2(Pa^{234})$, $y=11$ line, ratio 322; and $Ms_1(Ra^{228})$ and $UX_1(Th^{234})$, $y=14$ line, ratio $=118$. This sort of agreement of the ratios might be expected since the two groups of three beta-active nuclei occupy closely similar positrons with respect to the minimum of the energy surface. It is strange, however, that the agreement is so good since UX_2 and Ms_2 lie on different Sargent curves.[8] This seems to indicate that one group of three lies on one Sargent curve, the others on the other one. The beta-active Pa^{233} must give U^{233} which is undoubtedly an alpha-emitting nucleus. There seems to be no reliable way to estimate the half-life of U^{233} and other alpha-emitting nuclei of the $4n+1$ chain since the constants for their Geiger-Nuttall line are not known. In general, however, for two alpha-emitting nuclei of the same Z but differing in mass by 2 units the half-life of the heavier one is greater by a factor of about 10^4. Comparing U^{233} with $AcU(U^{235})$would thus give a half-life of the order of 10^5 years for U^{233}.

For ${}_{93}EkaRe$ the rule gives mass numbers of 239, 237 and 235. 235 can be excluded because of the existence of U^{235}. $EkaRe^{239}$ is analogous to Pa^{233} in being the nucleus formed by the beta-active U^{239} which is produced in the capture of slow neutrons by U^{238}. It lies on the $y=10$ line. From the near equality of the half-lives of U^{239} and Th^{233} we may infer that $EkaRe^{239}$ is probably also beta-active and with a half-life of about one month. This estimate is not in disagreement with the results of the experiments which have been made in the attempts to find an activity[9] of $EkaRe^{239}$. The ${}_{94}EkaOs^{239}$ which thus results from the disintegration of ${}_{93}EkaRe^{239}$ is undoubtedly an alpha-active nucleus of long half-life. 237, another $4n+1$ number, is left for beta-stable ${}_{93}EkaRe$. If $EkaRe^{237}$ is beta-stable, then U^{237} is presumably beta-active. This is plausible since it is the analog of beta-active $UY(Th^{231})$. U^{237} could probably be produced from U^{238} by fast neutrons as Th^{231} was produced by Nishina *et al.*, although the yield would be much smaller because of the greater probability for fission.[10]

For ${}_{95}EkaIr$ the rule gives 245, 243, and 241 as possible mass numbers, 243 lying on the $y=8$ line with Pa^{231} and $EkaRe^{237}$.

Applying the rule with the interpolated values of 214 for ${}_{84}Po$ and 220 for ${}_{86}Em$ gives 215, 213, and 211 as masses for ${}_{85}EkaI$ and 221, 219, and 217 for ${}_{87}EkaCs$. 221 and 215 lie on the $y=10$ line. To the left of them on this line lie stable ${}_{83}Bi^{209}$ and ${}_{81}Tl^{203}$, to the right are beta- and alpha-active ${}_{89}Ac^{227}$ and beta-active ${}_{91}Pa^{233}$. One might guess that ${}_{87}EkaCs^{221}$ and ${}_{85}EkaI^{215}$ would be beta-stable, but with a predominant alpha-activity whether beta-stable or not.

The rule gives 101, 99, and 97 for ${}_{43}Ma$. All three of these nuclei of Ma would be isobaric with a known stable isotope of either ${}_{42}Mo$ or ${}_{44}Ru$ so that there is presumably no stable isotope of Ma, as has been pointed out before. For ${}_{61}Il$ the rule gives 151, 149, 147. ${}_{62}Sm^{149}$ and ${}_{62}Sm^{147}$ are known so that 151 is the only likely mass number of Il.

The Fission of Transuranic Nuclei

Bohr and Wheeler[3] have shown that fission of a nucleus can occur when it attains a certain critical deformation. Only those nuclei which

TABLE I.

Z	90	91	92	93	94	95	96
A_{crit}	226.0	231.0	236.1	241.2	246.3	251.6	256.9
A_{least}	226	231	236	241	246	251	256
E_{min}(Mev)	0.2	0.2	0.2	0.2	0.1	0.1	0.0
A_{max}	232	231	238	237	244	243	250

[7] Meitner, Hahn and Strassmann, Zeits. f. Physik **109**, 538 (1938).

[8] J. S. Marshall, Proc. Roy. Soc. **173**, 391 (1939).

[9] E. Segrè, Phys. Rev. **55**, 1104 (1939); J. W. Irvine, Phys. Rev. **55**, 1105 (1939).

[10] Nishina, Yasaki, Ezoe, Kimura, and Ikawa, Nature **144**, 517 (1939).

have nearly the full energy necessary for such deformation will have a high probability of undergoing fission. The available energy is the sum of the energy of binding and of the kinetic energy of a captured neutron. It amounts to 5.2 Mev for the capture of thermal neutrons by the heaviest alpha-active nuclei of Th and U and might be expected to be nearly the same for similar atoms of higher atomic number. (See Table III of Bohr and Wheeler.[3])

From Fig. 4 of Bohr and Wheeler's paper it may be inferred that all nuclei for which $Z^2/A < 35.87$ will require less than this energy of 5.2 Mev for the critical deformation and can, therefore, undergo fission with thermal neutrons. From $Z^2/A_{\text{crit}} = 35.87$ are computed the values of A_{crit} of Table I. In general, for any bombarded nucleus whose A is less than $A_{\text{crit}} - 1$, fission will be produced by thermal neutrons and all faster ones. For $A > A_{\text{crit}} - 1$ fission will not be produced by thermal neutrons unless the nucleus is one containing an odd number of neutrons. For this latter sort of nucleus the energy of binding of an additional neutron is about 1.0 Mev higher, so that fission may occur. In accord with this there is given in the third row of Table I for each Z a value of A_{least}, the lowest value of A for a target nucleus with which fission could not be produced by thermal neutrons. In the fourth row is the corresponding value of E_{min}, the estimated minimum kinetic energy that a neutron would have to have in order to produce fission of the A_{least} nucleus. In the last row of Table I are the masses of the heaviest beta-stable nuclei as known or as obtained in the foregoing discussion.

Since the probability of fission increases rapidly with increasing kinetic energy of the nucleus being captured, it is clear that the nuclei which undergo fission with slow neutrons are the ones which will give large yields of fission with fast ones of assorted energies. All of the A_{least} nuclei would thus be fairly susceptible to fission by neutrons of moderate energy (<0.5 Mev). Nuclei of masses greater by 1 unit would give fission with thermal neutrons because of the greater energy of binding of a neutron into nuclei which originally contain an odd number of them. Those of masses greater by 2 units will require an additional 0.5 Mev of kinetic energy of the impinging neutrons for fission to be possible and will therefore be relatively stable. Since the masses of the A_{least} nuclei are all greater than the highest expectable masses of stable nuclei for the transuranic elements, these would be destroyed very effectively by exposure to neutrons. The difference between A_{least} and A_{max} is for each transuranic element so great that the general conclusion is valid irrespective of minor inaccuracies in the difficult estimation of the critical energy for fission. Recent results of the Westinghouse group[11] indicate that the critical energy is somewhat lower for U^{238} and Th^{232} than estimated by Bohr and Wheeler so that the fission of transuranic elements is even more probable.

High probability of fission of U^{236} is also indicated by the value of A_{least} for U.

In the foregoing discussion it is implied that the nucleus must have the energy of the critical deformation for fission to be important. It is to be expected, however, that nonclassical penetration of the potential barrier will give some fission at lower energies. There must be a small but finite probability for spontaneous fission of all nuclei which can divide exothermically. The failure to observe any such effect with U^{238}, U^{235} or U^{234} means that its probability must be very slight in view of their long half-lives. I am indebted to my colleague, J. A. Wheeler, for pointing out that such spontaneous fission may, nevertheless, offer an alternative way of accounting for the absence of the transuranic atoms. The same lowering of the critical energy which makes the transuranic atoms sensitive for fission by slow neutrons will also greatly increase this probability for spontaneous fission. Whether it becomes great enough to be of importance for any of the nuclei here considered cannot now be said with assurance.[12] The probability for fission is undoubtedly greatest for the nuclei with the largest values of Z^2/A. Of all of those discussed in this paper as being obtainable by possible experiments the hypothetical $_{94}EkaOs^{239}$ would be the one most likely to give spontaneous fission. For it, $x = (Z^2/A)/(Z^2/A)_{\text{limiting}}$ has a value of

[11] Haxby, Shoupp, Stephens, Wells and Goldhaber, Phys. Rev., this issue, p. 1088. The fission threshold for Th is about 1.1 Mev instead of the predicted 1.7 Mev.

[12] See the discussion by Bohr and Wheeler, reference 3, p. 435, and a further discussion in a paper by them which is to appear later.

0.772 corresponding to a critical energy of about 4 Mev. $_{92}U^{232}$ and $_{93}U^{237}$ come next, both having values of x of 0.763 which corresponds to 4.4 Mev.

The Production of U^{235} from U^{238}

The occurrence in nature of U^{235}, which is highly susceptible to fission, seems to be incompatible with the hypothesis that the transuranic atoms have been eliminated by this process. To see that there is no contradiction it is necessary to consider the capture of neutrons which will take place. Those captured by U^{238} form U^{239} which emit beta-particles to give $EkaRe^{239}$. As discussed in a foregoing section this also is probably beta-active and produces $EkaOs^{239}$ which would be expected to emit alpha-particles to give U^{235}. The $EkaOs^{239}$ is the analog of the missing U^{233} and would thus be expected to have a half-life short compared to that of U^{235}. It is not necessary to make detailed assumptions, for in any case it is most probable that the $EkaRe^{239}$ will somehow disintegrate to produce U^{235}. The concentration of U^{235} will be built up by this process and simultaneously lowered by fission of U^{235}. The relative concentrations, N_{238} and N_{235}, may be estimated by setting the rate of production of U^{239} nuclei equal to the rate of loss of U^{235} nuclei provided that we make the plausible assumption that the half-lives of the $_{93}EkaRe^{239}$ and possible $_{94}EkaOs^{239}$ are short enough compared to that of $_{92}U^{235}$ that no appreciable number of atoms will be lost by fission in these intermediate stages. The number of $_{92}U^{239}$ nuclei, and therefore of $_{92}U^{235}$ nuclei, produced per unit time by the above assumption will be proportional to $N_{238}\times\sigma_{r238}$. σ_{r238} is the averaged cross section for radiative capture of neutrons of different velocities by the U^{238} nuclei. Similarly, the rate of disappearance of U^{235} nuclei will be proportional to $N_{235}(\sigma_{r235}+\sigma_{f235})$, σ_{f235} being the cross-section for fission of U^{235}. The constant of proportionality involving the concentration and distribution in velocity of the neutrons will be the same for both. Setting these equal to each other gives

$$N_{235}/N_{238}=\sigma_{r238}/(\sigma_{r235}+\sigma_{f235}).$$

We may introduce the Γ's, the widths or probabilities of disintegration per unit time to get[13]

$$\frac{N_{235}}{N_{238}}=\frac{\Gamma_{r238}}{\Gamma_{r238}+\Gamma_{f238}+\Gamma_{n238}}\cdot\frac{\Gamma_{r235}+\Gamma_{f235}+\Gamma_{n235}}{\Gamma_{r235}+\Gamma_{f235}}\cdot\frac{R^2{}_{238}}{R^2{}_{235}}.$$

The R's are the radii of the two nuclei, approximately equal. Γ_{f235} and Γ_{n235} are of the same order of magnitude for energies of a few million volts and both much greater than Γ_{r235}. Γ_{r238} and Γ_{f238} are apparently considerably smaller than Γ_{n238} so that the conclusion is that N_{235}/N_{238} should be considerably less than unity, being somewhat greater than $\Gamma_{r238}/\Gamma_{n238}$. The abundance ratio U^{235}/U^{238} corrected to a time 2×10^9 years ago is 1 : 27 according to Nier.[14] This result is not incompatible with the assumed formation of U^{235} from U^{239}.

The $4n+1$ Radioactive Atoms

In the section on the beta-stable nuclei the probable properties of several of the nuclei of the missing $4n+1$ family have been discussed. It seems probable that the successive disintegrations would be as follows:

$$U^{237}\xrightarrow{\beta}EkaRe^{237}\xrightarrow{\alpha}Pa^{233}\xrightarrow{\beta}U^{233}\xrightarrow{\alpha}Th^{229}\xrightarrow{\alpha}Ra^{225}\xrightarrow{\beta}Ac^{225}$$
$$\xrightarrow{\alpha}EkaCs^{221}\xrightarrow{\alpha}EkaI^{217}\xrightarrow{\alpha}Bi^{213}\xrightarrow{\alpha}Tl^{209}\xrightarrow{\beta}Pb^{209}\xrightarrow{\beta}Bi^{209}.$$

Bi^{213} would probably show branching as do the other C products so that some nuclei would go as follows:

$$Bi^{213}\xrightarrow{\beta}Po^{213}\xrightarrow{\alpha}Pb^{209}\xrightarrow{\beta}Bi^{209}.$$

The foregoing $4n+1$ chain of disintegrations is shown by the heavier arrows of Fig. 2. Th^{233} does not appear in this chain but there is no more reason why it should do so than that the Ra^{227} and Pa^{232} which undoubtedly could be produced by capture of slow neutrons by Ra and Pa should appear in the natural Ac and Th chains. Insofar as the half-lives of the nuclei of the above chain can be inferred by analogies and their positions with respect to known nuclei it seems that no one of them should have a half-life comparable with the long half-lives of U, Th and AcU. The existence in nature of all of the heavy radioactive atoms apparently depends

[13] See the last paragraph of Section III of Bohr and Wheeler, reference 3.

[14] A. O. Nier, Phys. Rev. **55**, 153 (1939), especially Table IV.

upon the great magnitude of these three half-lives which have insured a continuing supply of the radioactive atoms. If there is any such nucleus at all in the $4n+1$ family it must be sought among the transuranic ancestors of U^{237}. $_{94}EkaOs^{241}$, the probable immediate ancestor of U^{237} might be expected to have a very long half-life ($y=9$, as for U^{235}). The absence of the $4n+1$ atoms thus becomes merely another aspect of the absence of transuranic atoms.

The assumed irradiation by neutrons would produce $4n+1$ atoms by their capture by Th^{232}. At first glance it seems peculiar that the abundance of their end product, Bi^{209}, is so low[15] compared to that of Pb^{207}. Bi^{209}, however, would have been partially eliminated and brought into equilibrium with Pb^{206}, Pb^{207}, and Pb^{208} in the assumed irradiation by neutrons, by reactions of the following cycle. $Pb^{206}+n\rightarrow Pb^{207}$, $Pb^{207}+n\rightarrow Pb^{208}$, $Pb^{208}+n\rightarrow Pb^{209}$, $Pb^{209}\rightarrow Bi^{209}+e^{-}$, $Bi^{209}+n\rightarrow Bi^{210}$(RaE), $Bi^{210}\rightarrow Po^{210}+e^{-}$, $Po^{210}\rightarrow Pb^{206}+\alpha$. It is not clear on this hypothesis why the abundance of Bi^{209} is only about 0.01 that of Pb^{207}. The spontaneous fission of $_{94}EkaOs^{241}$, however, would account for the scarcity of its remote descendant, Bi^{209}.

U^{236} and U^{232}

At first glance the absence of U^{236} seems unusual. Sm^{146} is the only other such even-even nucleus in the whole periodic table which is absent when both the even-even nuclei of mass greater by 2 units and less by 2 units exist. The bracketing of U^{236} by U^{238} and U^{234} is, however, perhaps not of any great significance since the latter is present only because of its transient equilibrium with U^{238}. The half-life of U^{236} may be estimated to be 10^5 yr. U^{236}, even if originally present in quantities comparable with that of U^{238}, would have disappeared if the age of the rocks of the earth is of the order of 2×10^9 yr. Any minerals in which it was originally present should now contain equivalent quantities of Th. Its absence in nature is also merely one aspect of the absence of its possible transuranic ancestors.

U^{232} would be expected to be a beta-stable nucleus which could exist because of the existence of Pa^{231}. There is no place in the list of isotopes of the stable elements for $Z>8$ where the even-even nucleus of mass one unit greater than the lightest stable isotope of the preceding element does not exist. U^{232} might be expected to have a half-life of about 2 yr. Its absence in nature is obvious since it does not occur in the Th chain. It would probably be produced from Pa^{232} formed by capture of neutrons by Pa^{231}.

Discussion of the Fission Hypothesis

As shown in the foregoing discussion the hypothesis of fission, either spontaneous or induced by neutrons, does account for the absence of the missing atoms. A steeper rise of the energy surface beyond $A=238$, like the one near $A=211$ which causes the short half-lives of the nuclei for $Z=84$ and 86, would, however, account for all of the facts equally well, except for the above-mentioned scarcity of Bi^{209}. If the distribution of nuclear species was once one of an equilibrium at a high temperature the relative abundances must have depended upon the energies and statistical weights of the different nuclei. There is no obvious reason why the $4n+1$ nuclei should have been less abundant than the $4n+3$ nuclei. If the absence of the $4n+1$ *radioactive* nuclei, transuranic and otherwise, is to be attributed merely to shortness of all half-lives, they should appear as a larger abundance of Bi^{209}, contrary to the fact. The very existence of all the heavy atoms is something of a puzzle, but it seems unlikely that any general hypothesis concerning their origin would favor $4n+3$ atoms markedly in comparison with $4n+1$ atoms.

More definite experimental evidence concerning the properties of $EkaRe^{239}$ and particularly of the possible $EkaOs^{239}$ will show whether there is a new rise in the energy surface beyond $Z=92$ which would lead to short half-lives for emission of alpha-particles. If one accepts the conclusion that $EkaRe^{239}$ is a beta-active nucleus, then the results of Segrè[9] indicate that $EkaOs^{239}$ does have a long half-life, excluding an abrupt rise of the energy surface. Segrè was unable to find any additional alpha-activity in a sample of U which had been exposed to strong intermittent bombardment by neutrons for over two years. As mentioned above, this nucleus seems to be the most promising one to scrutinize more carefully in a search for possible spontaneous fission.

[15] F. W. Aston, *Mass Spectra and Isotopes* (1933), especially Fig. 36 on p. 185.