CHAPTER I

THE SYSTEMATICS OF STABLE NUCLEI

1.1 Empirical definition of stability

The concept of nuclear stability requires careful definition. Formally the matter is simple enough: a nucleus is said to be stable when it is not subject to spontaneous disintegration; in practice, however, it is not always easy to decide the question one way or the other by direct experiment.

For nearly forty years α- and β-disintegration were the only types of spontaneous disintegration known to occur, but the study of 'artificially' produced species during less than half that time has added positron emission and orbital electron capture to this list—and the more recent discovery of neutron-induced fission has been followed by the recognition that fission also occurs as a spontaneous process with the heaviest nuclei (Petrzhak and Flerov, 1940). Here we may note a problem in conventional nomenclature. We have to envisage the possibility of types of spontaneous transformation in which nuclei divide into two fragments between which the mass is shared in any proportion: obviously the term fission might serve to describe all such processes (less well, admittedly, for electron and positron emission) but it has acquired a particular meaning. In actual fact the acute problem has not yet arisen; we speak of α-disintegration when the lighter fragment is the helium nucleus, we should certainly speak of spontaneous proton emission if a hydrogen nucleus were involved, but we have also to be prepared in theory to consider other light nuclei as possible products of transformation. It may conceivably be held that the characteristic of 'fission' is that any spontaneously fissionable nucleus may divide in a number of different ways, but in the limiting case clearly this number will not be greater than one, and the distinction will disappear. Evidently there is a problem in nomenclature involved, though it is as yet an academic one.

It has been implied that it is not always easy to decide by direct experiment whether a given element is stable or unstable, that is whether or not any isotopic constituent of the element is unstable

as it occurs under terrestrial conditions. Since the present chapter deals with the 'stable' nuclei, defined for our immediate purpose as those nuclei not known to be unstable, it will be well to examine this statement further so that the degree of uncertainty in any case can be appreciated. We shall see that what is important, for all possible types of instability, is the dependence of disintegration probability upon available energy—and also, in certain circumstances, the rate of loss of energy with distance, for the disintegration particles, in the medium in which they are to be detected.

In respect of α-disintegration we may note that no case is known, with the 'classical' radioelements, in which less than 4 MeV. of energy is liberated. This result may well reflect a definite discontinuity in nuclear properties, considered as a function of nuclear charge, for a charge number in the neighbourhood of 82 (see p. 46), but it certainly bears witness to a practical limitation which is undoubtedly effective. The half-value period of thorium (α-disintegration energy 4·05 MeV.) is $1{\cdot}40 \times 10^{10}$ years; if the α-disintegration energy had been 3·3 MeV. or less the half-value period would have been at least 10^{15} years. In that case the natural radioactivity of thorium would probably have escaped recognition hitherto. Similarly, it can be stated that it is impracticable at present to decide, at least by direct experiment, whether or not naturally occurring lead or bismuth are α-active, except in respect of disintegration energies greater than about 3 MeV.† For disintegration energies less than this amount the rate of disintegration of these elements would be too slow for certain detection. For elements of smaller nuclear charge, however, less uncertainty in respect of energy remains; disintegration probabilities increase, for a given energy, as the nuclear charge decreases (p. 74) (the active isotope of samarium, $^{147}_{62}$Sm, has a half-value period of $1{\cdot}1 \times 10^{11}$ years (Cuer and Lattes, 1946; Picciotto, 1949) and a disintegration energy of 2·24 MeV. (Jesse and Sadauskis, 1949)), on the other hand the difficulty of detecting α-particles of small energy emitted by feebly active preparations eventually asserts itself, so that the situation in respect of minimum detectable instability does not

† Faraggi and Berthelot (1951) have recently reported the observation of α-particles of energy 3·15 MeV. which they attribute to the disintegration of $^{209}_{83}$Bi with half-value period $2{\cdot}7 \times 10^{17}$ years.

improve indefinitely as the atomic number of the element concerned is reduced to the limit. Direct experiment has been unable to establish the stability of the common isotope of beryllium ($^{9}_{4}$Be) beyond a possible half-value period of 10^{14} years, assuming that the energy of α-emission might be as small as 0·1 MeV.† Probably the most sensitive method of investigation of general applicability is the method which employs the newly developed fine-grain emulsions for the detection of the emitted particles, but even with these emulsions it is difficult to recognize with certainty the tracks of α-particles of less than about 0·6 MeV. energy, that is, of less than 4 mm. range in standard air. As recently employed, this method has yielded minimum lifetimes of the order of 10^{16} years for certain medium-heavy elements.‡

The problem of feeble β-activities is not complicated by any important variation of lifetime with nuclear charge, for a given energy of disintegration. In another respect, however, the position is more complicated than with α-disintegration. In the case of β-disintegration, disintegration probabilities are not represented by a single-valued function of the energy, as, to a first approximation, α-disintegration probabilities are, for a specified nuclear charge. Depending upon the details of the transformation (cf. p. 93), β-disintegrations are classified as 'allowed', 'once-forbidden', 'twice-forbidden', etc., and disintegration probabilities may vary by a factor of 10^{2} or 10^{3}, for a given energy, for a change in type of disintegration represented by the difference between one class and the next. If it were not for this effect there would, in fact, be no outstanding problem concerning possible β-activities of long lifetime; if all disintegrations were 'allowed' the half-value period for any activity having a characteristic energy of greater than, say, 0·01 MeV. would be measured in thousands of years, at the most. Clearly, no species of such short lifetime could exist in appreciable concentration on the earth to-day, unless it were continuously being produced from a much longer-lived parent which then of necessity would be heavy-particle active. However, all β-disintegrations are not of the 'allowed' type, and within recent years β-activity

† Rayleigh, 1933; Evans and Henderson, 1933; Gans, Harkins and Newson, 1933; Libby, 1934.

‡ Broda, 1947; Jenkner and Broda, 1949; Bestenreiner and Broda, 1949*a*, 1949*b*.

of long lifetime has been established for $^{115}_{49}$In (Martell and Libby, 1950), $^{176}_{71}$Lu (Heyden and Wefelmeier, 1938; Libby, 1939) and $^{187}_{75}$Re (Naldrett and Libby, 1948*a*, 1948*b*; Sugarman and Richter, 1948), and the previously recognized β-activities of potassium and rubidium have been assigned to the common species $^{40}_{19}$K (v. Hevesy, 1935; Smythe and Hemmendinger, 1937) and $^{87}_{37}$Rb (Mattauch, 1937; Hahn, Strassmann and Walling, 1937). Moreover, there have been reports that $^{145}_{60}$Nd† is also β-active (see, however, p. 17), and the recently discovered $^{138}_{57}$La (Inghram, Hayden and Hess, 1947*b*, 1947*c*) and $^{50}_{23}$V (Hess and Inghram, 1949; Leland, 1949*b*) can hardly be β-stable (Pringle, Standil and Roulston, 1950; Pringle, Standil, Taylor and Fryer, 1951). As an indication of the degree of uncertainty in respect of energies and lifetimes which may still persist in respect of a small number of species, possibly subject to highly forbidden β-disintegrations such as these, it may be stated that the maximum energy of the β-particles emitted by $^{187}_{75}$Re is 0·043 MeV. and that the half-value period of this species (of natural abundance 62·9% in ordinary rhenium) is roughly 4×10^{12} years. It is not impossible, therefore, that one or two comparatively rare species, at present classified as stable, may be undetected β-emitters of small disintegration energy—say 0·01 MeV. or less—and of lifetime no greater than 10^9 years.

What has been said concerning β-disintegration applies equally to positron emission, though, up to date, no naturally occurring positron emitter has been discovered. The case of orbital electron capture, however, is less hypothetical, and it raises new problems. It has been established that $^{176}_{71}$Lu undergoes transformation of this type as an alternative to β-disintegration (Flammersfeld, 1947) and the indications are that $^{40}_{19}$K exhibits similar branching,‡ and that $^{138}_{57}$La also is capture-active (vide supra). From the point of view of experiment the main problem posed by the occurrence of orbital electron capture is that the primary radiation is not a particle radiation. Methods of detection, therefore, are generally insensitive. Moreover, with the lighter elements the primary radiation is

† Libby, 1934; Feather, 1945; Ballou, 1948; Jha, 1950; Wapstra, 1952.

‡ v. Weizsäcker, 1937; Thompson and Rowlands, 1943; Bleuler and Gabriel, 1947; Stout, 1949; Sawyer and Wiedenbeck, 1949, 1950; Bell and Cassidy, 1950; Spiers, 1950; Ceccarelli, Quareni and Rostagni, 1950; Inghram, Brown, Patterson and Hess, 1950; Good, 1951; Colgate, 1951.

relatively easily absorbed in the source material—or in the air: the K X-radiation from potassium, for example, is almost completely absorbed in a few centimetres of standard air. It is true that a nuclear γ-radiation may follow the emission of the 'primary' fluorescent X-radiation in an appreciable fraction of the transformations with some capture-active species, making possible the recognition of a number of activities which would otherwise go undetected,† but it is not possible to rely on such a favourable circumstance in all cases. When this feature is absent it can almost be said that any of the lighter capture-active species which is sufficiently long-lived to exist in detectable amount upon the earth is also so feebly active as to appear stable under direct experimental examination. Even with the heaviest species, for capture activity to be recognized with certainty, the lifetime should not be greater than 10^{12} years. Further it must be noted that capture processes may remain undetected even through the energy of instability is not very small: if there is no available state of excitation of the product nucleus (which might be exhibited in γ-ray emission) the whole of the excess energy is carried by the unobservable neutrino.

One final remark concerning limits of detection is called for here. The figures which have so far been given refer to the recognition of instability by the detection of the radiation emitted from disintegrating nuclei. In certain cases geochemical methods provide a more sensitive test. Such methods depend upon the accumulation of a stable end-product of disintegration during geological time, and they are particularly suitable when this end-product is an inert gas. An extreme case is represented by the mass analysis of the xenon occluded in an old tellurium mineral which has established the stability of $^{130}_{52}$Te against the simultaneous emission of two β-particles as far as a lower limiting lifetime of 8×10^{19} years (Inghram and Reynolds, 1949). Obviously this conclusion depends upon a knowledge of the age of the mineral concerned, and upon the assumption of complete retention of occluded gas during the time for which the mineral has existed in the solid phase. A less extreme case, but one less open to doubt concerning possible disappearance of the end-product of disintegration, is provided by the chemical

† The K-capture transformation of the artificially produced $^{7}_{4}$Be is an extreme example of this.

estimation of indium in an old cassiterite, which has led to the conclusion that $^{115}_{50}$Sn is stable against capture transformation unless the lifetime for this process is greater than 5×10^{12} years (Ahrens, 1948). In respect of double β-disintegration, mentioned above as a possibility for $^{130}_{52}$Te (see also § 1.2, below), it might be pointed out that the availability of the coincidence method of detection in such cases increases the sensitivity of the direct search for possible radioactivity. Thus it has been concluded that the lifetime of $^{124}_{50}$Sn against double β-decay is of the order 10^{17} years or longer (Fireman, 1949; Kalkstein and Libby, 1952)—representing a sensitivity at least three powers of ten better than could have been achieved by the usual method of single counting (see also Lawson, 1951).

1.2. Neighbouring stable isobars

The problem of the relative stability of isobaric nuclear species (that is, nuclear species characterized by a single value of the mass number, A) is closely linked with the problem of capture transformation which we have just considered. For that reason it may be discussed at this stage, although the stability rule involved was not one of the earliest to be enunciated when the results of the mass analysis of the elements first came under review. The rule is, briefly, that pairs of isobaric stable nuclei differ in charge number (Z) very much more frequently by two units than by one (Meitner, 1926). Current tables of 'existing' stable nuclei contain sixty-five examples† of the former relation and only two of the latter (it is assumed that the free neutron is β-active (Snell, Pleasanton and McCord, 1950; Robson, 1950, 1951). The two pairs of neighbouring isobars, for each of which both members are currently regarded as stable, are $^{113}_{48}$Cd and $^{113}_{49}$In, and $^{123}_{51}$Sb and $^{123}_{52}$Te. It is probably not without significance that these species lie so close together in the sequence of the elements.

The reason for the infrequent occurrence of neighbouring stable isobars is not far to seek. Let us consider the neutral atoms of two neighbouring isobars, $\binom{A}{Z}$ and $\binom{A}{Z+1}$. If the masses of these two

† Eight of these pairs may further be grouped as four triads in each of which the three charge numbers stand in arithmetic progression with common difference two units. The species concerned are: $^{96}_{40}$Zr, $^{96}_{42}$Mo and $^{96}_{44}$Ru; $^{124}_{50}$Sn, $^{124}_{52}$Te and $^{124}_{54}$Xe; $^{130}_{52}$Te, $^{130}_{54}$Xe and $^{130}_{56}$Ba; and $^{136}_{54}$Xe, $^{136}_{56}$Ba and $^{136}_{58}$Ce.

atoms are not identical one or other atom must have the greater mass. Let us suppose, to begin with, that the neutral atom $\binom{A}{Z}$ is heavier than the neutral atom $\binom{A}{Z+1}$. Then, if it is assumed that no uncharged particle of finite rest-mass is emitted from the nucleus in the process of β-disintegration (that is, in contemporary theoretical phraseology, that the rest-mass of the neutrino is zero), and if the mass-difference between the two neutral atoms is greater than a small quantity $W_v(Z+1)/c^2$ which we shall presently define, it must be energetically possible for the species $\binom{A}{Z}$ to transform spontaneously into the species $\binom{A}{Z+1}$ by β-emission. In this case, clearly, both species cannot be stable.

Consider now the other alternative, namely that the mass of the neutral atom $\binom{A}{Z+1}$ is greater than the mass of the neutral atom $\binom{A}{Z}$. Then, making the same assumption concerning the mass of any hypothetical uncharged particle emitted in the process, if the difference in mass between the neutral atoms is greater than $W_K(Z)/c^2$, where $W_K(Z)$ is the K-ionization energy for an originally neutral atom of charge number Z, K-electron capture will be energetically possible for the species $\binom{A}{Z+1}$, and the assumption that the neighbouring isobars $\binom{A}{Z}$ and $\binom{A}{Z+1}$ are both essentially stable is obviously untenable. Again, in principle, at least (Marshak, 1942), a similar statement relates to transformation by capture of a less tightly bound extranuclear electron,† if the appropriate ionization energy, $W_L(Z)$, $W_M(Z)$, ..., is substituted for $W_K(Z)$.

We come to this conclusion, therefore (always assuming zero rest-mass for the neutrino, a result to which the experimental evidence steadily approximates (Hanna and Pontecorvo, 1949; Curran, Angus and Cockroft, 1949; Kofoed-Hansen, 1947, 1951)),

† L-electron capture has been experimentally detected for ${}^{37}_{18}\mathrm{A}$ by Pontecorvo, Kirkwood and Hanna (1949).

that two neighbouring isobars cannot both be stable unless

$$-W_v(Z+1)/c^2 < M\begin{pmatrix} A \\ Z+1 \end{pmatrix} - M\begin{pmatrix} A \\ Z \end{pmatrix} < W_v(Z)/c^2.\dagger$$

Here $W_v(Z)$ is the first ('valency') ionization energy and $M\begin{pmatrix} A \\ Z \end{pmatrix}$ is the mass of the neutral atom $\begin{pmatrix} A \\ Z \end{pmatrix}$, c being the velocity of light.

Since $W_v(Z)/c^2$ is of the order of 10^{-8} mass unit for all Z, the condition for the absolute stability of neighbouring isobars of any mass number is that the masses of the neutral atoms shall be the same within this narrow margin. Since there is no fundamental reason for such near identity of mass, the masses of neutron and proton differing by $1{\cdot}4 \times 10^{-3}$ mass unit, any case in which the condition is satisfied must be a case of coincidence. It is extremely unlikely, then, that there should even be two such cases in the system of the stable elements of natural occurrence, and we are led to suppose that one or other of the species in each of the pairs already listed is in fact unstable, though its disintegration is evidently of a highly forbidden type. It may be noted that, but for their recognized activity, the long-lived species ${}^{40}_{19}$K, ${}^{87}_{37}$Rb, ${}^{115}_{49}$In, ${}^{176}_{71}$Lu and ${}^{187}_{75}$Re would form neighbouring stable-stable pairs with ${}^{40}_{20}$Ca (and ${}^{40}_{18}$A), ${}^{87}_{38}$Sr, ${}^{115}_{50}$Sn, ${}^{176}_{72}$Hf (and ${}^{176}_{70}$Yb) and ${}^{187}_{76}$Os. In another section (p. 24) evidence will be presented which suggests that ${}^{113}_{48}$Cd and ${}^{123}_{51}$Sb are probably the unstable (β-active) members of the anomalous pairs.

At this point a somewhat academic remark is worth making, particularly because of its relevance for a later chapter (p. 118). It is that the conditions of stability of bare nuclei are different from those which have just been given for neutral atoms. Thus a little consideration will show that for the bare nucleus $\begin{pmatrix} A \\ Z \end{pmatrix}$ β-disintegration to form the nucleus $\begin{pmatrix} A \\ Z+1 \end{pmatrix}$ is possible only when

$$M\begin{pmatrix} A \\ Z \end{pmatrix} - M\begin{pmatrix} A \\ Z+1 \end{pmatrix} > \{W(Z+1) - W(Z)\}/c^2,$$

† It will be obvious that the limits would be further narrowed, by the replacement of $-W_v(Z+1)/c^2$ by zero, if β-disintegration into a bound (valency) state were regarded as possible (Daudel, Jean and Lecoin, 1947; Jean, 1948; Sherk, 1949; Ivanenko and Lebeder, 1950).

$W(Z)$ being the total energy necessary to ionize the atom completely—a quantity of the order of $5{\cdot}5\,W_K(Z)$ (Allard, 1948; Foldy, 1951). Similarly, for positron emission to be possible from the bare nucleus $\binom{A}{Z+1}$, the masses of the neutral atoms must satisfy the inequality

$$M\binom{A}{Z+1} - M\binom{A}{Z} > 2m - \{W(Z+1) - W(Z)\}/c^2,$$

whereas, when the initial and final atoms are themselves neutral, the corresponding condition is

$$M\binom{A}{Z+1} - M\binom{A}{Z} > 2m,$$

m being the electronic mass.

Obviously, the distinction here made presupposes the reality of some kind of coupling between the energy states of the nucleus and of the outer atom; that supposition being granted we see that total ionization of an atom may, in the marginal case, suppress the radioactivity of a species which is normally β-active† or bring to light the latent positron activity of a species which appears positron-stable in the neutral state. It is hardly necessary to point out that the limits for the incidence of this effect are narrow ones, being of the order of $10^{-7}\,Z$ mass unit, or about $150Z$ eV., but it seems that at least one example of its operation has been recognized.‡ As regards the capture process, there is no sense, of course, in speaking of K-electron capture by a bare nucleus, but it will appear from what has previously been said that this type of transformation is energetically most probable when the initial atom is already completely ionized except for a single electron in the K shell—and a similar remark holds for L-, M-, ... electron capture, as the case may be. Ultimately, capture of a free electron by a bare nucleus is seen as energetically most favoured of all.

Having established a satisfactory explanation of the rarity of pairs of neighbouring 'stable' isobars, it is interesting to return to

† As before we are neglecting β-disintegration into bound states. Such disintegration cannot similarly be suppressed by ionization of the atom.

‡ Indications are that the energies of β-disintegration for the predominant modes of $^{227}_{89}$Ac and $^{228}_{88}$MsTh_1 are less than $150Z$ eV ($\sim$13 keV) in each case.

the question of the frequency of occurrence of pairs of apparently stable isobaric species for which the difference of charge number is two. For it will be recognized that the type of analysis already given is equally valid for this case, if double β-disintegration and double electron capture are accepted as possible unit events. On this basis, then, one or other member of each such pair of isobars must in the last resort be regarded as essentially unstable. The fact that no example of a double process—either double β-emission or double electron capture—has with certainty been established (experimental limits of stability have already been given for two species, see p. 5)† enforces the conclusion that the disintegration constant for such a process is always very small (say, $< 10^{-23}$ sec.$^{-1}$) even when the available energy is considerable (say, several MeV.). This conclusion can be reconciled with current theory (Touschek, 1948; Daudel and Jean, 1949) without much difficulty.

One other conclusion can clearly be drawn from the experimental facts. It is that if $\binom{A}{Z}$ and $\binom{A}{Z+2}$ are two 'stable' isobars differing in charge number by two, then the 'unobserved' intermediate species $\binom{A}{Z+1}$ must be essentially unstable in respect both of β-disintegration and of electron capture. If it were not unstable in respect of either transformation, then obviously the rule regarding neighbouring isobars would be violated for one pairing or the other, that is, either for the pairing $\binom{A}{Z}$, $\binom{A}{Z+1}$ or for the pairing $\binom{A}{Z+1}$, $\binom{A}{Z+2}$. In fact the predicted branching disintegration of these unstable intermediate species has so far escaped detection in the majority of cases (Feather, 1948*a*): clearly in each of these cases one disintegration mode is much more probable than the other.

1.3. The distinction between even and odd

The sixty-one‡ values of A, for which pairs of stable isobars differing by two units in Z are known to exist, are all even values—and the values of Z belonging to these paired species are also,

† Levine, Ghiorso and Seaborg (1950) have recently shown that the half-value period for the process $^{238}_{92}\mathrm{U} \xrightarrow{2\beta} {}^{238}_{94}\mathrm{Pu}$ is greater than 6×10^{18} years.

‡ See footnote, p. 6.