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#### Energy and Stability Related to Composition of Atomic Nuclei

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Examination of the postulates of Heisenberg concerning the energy of binding of neutrons and protons to atomic nuclei shows that factors not included in the primary considerations are of appreciable effect in determining nuclear composition and stability, and result in deviations of a periodic character from the requirements of the theory. Nevertheless, the formulation of binding energies as linear functions of the neutron-proton ratio of the nucleus is adequate as a rough approximation over a large range of elements, as is also the proportionality assumed between the radius and the cube root of the mass number. For evaluation of the various constants, it is assumed that the boundary of stability in emission of beta-particles should be centrally placed with respect to existing nuclear species. Reasons supporting this placement, and the manner of adjustment of the alpha-boundary, for which the distribution of nuclei alone is not an adequate guide, are outlined. The application in various fields of nuclear physics and chemistry of the constants thus determined is discussed and illustrated. The nuclear radii resulting in these calculations agree excellently with those found by other methods. An equation for the energy of formation of nuclei from neutrons and protons is obtained and found to hold, within experimental error, for all elements above 24Cr for which

HEISENBERG, in recent papers,<sup>1</sup> discusses the properties of atomic nuclei assumed to consist entirely of neutrons and protons. Among other results, he obtains approximations to the forms of the stability-composition relations in alpha- and beta-disintegrations. This phase of his analysis is based upon assumptions concerning the interaction of single neutrons and

data are available for comparison. Calculated energies of spontaneously emitted alpha- and beta-particles and energies of capture of various particles by light nuclei, are in rough agreement with known values. Energies in nuclear collision processes are calculated with fair average agreement with experimental values, and the indications of the theory regarding critical energies and nuclear composition in such reactions discussed. Specific characteristics are not faithfully predicted in the various nuclear changes above, but the average accuracy of calculated energies is superior, for all but the light elements, to that obtainable from present values of atomic masses. In addition, these calculations are possible in cases where the masses are not yet measured. Spontaneous emission of protons from any existing nuclei is indicated to be impossible. A chart is given showing in what region of nuclear composition this and other similar changes might occur. The alpha-disintegration is considered to be possible in elements as low as 51Sb. The possibility of beta-changes, or their reverse, in certain isobaric pairs among the "non-radioactive" elements is pointed out. The "end" of the periodic system and the limiting spread in mass numbers of isotopes as influenced by the various types of nuclear instability are also discussed.

protons with nuclei of various compositions, which may be cast in algebraic form as follows:

$$\overline{E}_N = K_1 + K_2 N/Z, \tag{1}$$

$$\overline{E}_P = k_1 + k_2 N/Z. \tag{2}$$

 $\overline{E}_N$  and  $\overline{E}_P$ , to be referred to as the binding energy of the neutron and the proton, designate the difference in energy between the highest point of the potential-distance curve (compare Fig. 3) and the lowest level of the particle when

<sup>&</sup>lt;sup>1</sup>Heisenberg, I, Zeits. f. Physik 77, 1 (1932); II, ibid. 78, 156 (1932); III, ibid. 80, 587 (1933).

combined in a nucleus synthesizable from N neutrons and Z protons.<sup>2</sup> The "radius" of the nucleus, designated by  $R_0$  and defined later, is assumed to be given by

$$R_0 = r_0 M^{1/3}, \tag{3}$$

where  $r_0$  is a constant.

Making these approximations, and the assumption that energy is conserved (statistically at least) in all processes considered, Heisenberg shows that the condition for zero energy in a beta-disintegration is

$$N/Z = C_1 + C_2 Z/M^{1/3}.$$
 (4)

Similarly, the limiting condition for stability with respect to alpha-emission is

$$N/Z = c_1 + c_2 Z/M^{1/3}.$$
 (5)

If  $E_N^{\circ}$  and  $E_{\alpha}^{\circ}$  represent the energy evolved in the formation from protons and electrons of a neutron and an alpha-particle, respectively, and e is the charge of the electron, the constants in (4) and (5) have the values

$$C_{1} = (K_{1} - k_{1} + E_{N}^{\circ})/(k_{2} - K_{2}),$$

$$C_{2} = e^{2}/r_{0}(k_{2} - K_{2}),$$

$$c_{1} = (E_{\alpha}^{\circ}/2 - E_{N}^{\circ} - k_{1} - K_{1})/(k_{2} + K_{2})$$
and
$$c_{2} = e^{2}/r_{0}(k_{2} + K_{2}).$$

Assuming that the upper limit of N/Z in existing nuclear species is fixed by (4) and the lower limit by (5), Heisenberg plots these extreme values against Z. Curves of the form of Eqs. (4) and (5) with the constants C and c empirically chosen delineate roughly the field within which the known nuclei lie, and to that extent support the theory.

Further support of the original assumptions, including a check of the magnitudes of the constants, is none the less desirable. Conversely, if the assumptions can be verified, and the stability curves correctly placed, the determination of the magnitudes of the constants in (1), (2) and (3) becomes itself of value. A detailed examination of the subject has therefore been made with the objectives of testing the assumptions, fixing the constants, and applying the theory to various problems of nuclear physics and chemistry.

#### A TEST OF THE BASIC ASSUMPTIONS

If Eqs. (1), (2) and (3) are valid approximations, and if the energy, E, evolved in the formation of any nucleus from neutrons and protons is a function of N and Z alone (that is, constitutional or structural factors are absent or negligible), the following equations should also be approximately true:

$$(\partial E/\partial N)_{Z} = \overline{E}_{N} = K_{1} + K_{2}N/Z, \qquad (6)$$

$$(\partial^2 E/\partial Z \partial N) = -K_2 N/Z^2, \tag{7}$$

$$(\partial E/\partial Z)_N = \overline{E}_P - Ze^2/r_0 M^{1/3}$$
  
=  $k_1 + k_2 N/Z - Ze^2/r_0 M^{1/3}$ , (8)

$$(\partial^2 E/\partial N\partial Z) = k_2/Z + Ze^2/3r_0M^{4/3}.$$
 (9)

Equating the second derivatives in (7) and (9) gives a relation which may be cast in the form

$$N/Z = -k_2/K_2 - Z^2 e^2/3K_2 r_0 M^{4/3}.$$
 (10)

Setting the constant term  $-k_2/K_2=D_1$ , and  $-e^2/3K_2r_0=D_2$ , this becomes

$$N/Z = D_1 + D_2 Z^2 / M^{4/3}, \qquad (10a)$$

which is the equation of a straight line if N/Zand  $Z^2/M^{4/3}$  are taken as the variables. The extent to which Eq. (10) or (10a) is satisfied, and the original assumptions substantiated, may therefore be simply tested by plotting these quantities for existing atomic species against each other.

The features which are of particular interest in plots of this kind are brought out most clearly by using values of N and  $M^{4/3}$  obtained from the chemical atomic weight, A, of each element. This provides a sort of average of the variables to be plotted which gives most weight to the isotopes of greatest abundance, allows each element to be represented by a single point, and permits the inclusion of the elements whose isotopic composition is as yet unknown. This plot is shown in Fig. 1.

<sup>&</sup>lt;sup>2</sup> Departure is here made from Heisenberg's notation partly for mnemonic reasons and partly in order to obtain the final equations in terms of quantities which are independent of any theory as to the nature of the nucleus. Thus, the number of protons in the present theory is the atomic number, usually represented by Z. Similarly if Mis allowed to represent the mass number of a nuclear species, N is M-Z. In Heisenberg's notation  $N=n_1$ ,  $Z=n_2$  and M=n.



FIG. 1. Test of assumptions averaging isotopic masses by use of chemical atomic weights.

The points in Fig. 1 show deviations from the straight line representing their general trend which are roughly but distinctly periodic in character. This behavior is not surprising, since the probable existence of more or less sharply marked "periods" in the system of nuclei is already known. It was first pointed out by Harkins, and is clearly shown in his recent plots<sup>3</sup> of isotopic number<sup>4</sup> against atomic number. It is most obvious among the lighter elements, and, in the present plot at least, is not distinguishable in the elements of atomic number greater than about 60.

The existence of this periodicity indicates that some factor not included in Heisenberg's simple assumptions is operative to a significant extent in determining nuclear composition. Some sort of exclusion principle might explain the periodicity, or it may result from the effects of spin, leading to structures of the kind proposed by Latimer.<sup>5</sup> Where it is necessary to consider these effects they will be referred to loosely as "structural factors." It will be considered here that they are superimposed upon the more general interchange forces, and are of secondary

importance, so far as energy effects are concerned, in large nuclei.

It can, therefore, by no means be claimed that the points in this diagram are best represented by a straight line. Nevertheless, insofar as rough numerical approximation is concerned, a linear function is adequate. In Fig. 1, for example, with the exception of a few of the light elements, deviations from the line are seldom more than 10 percent in N/Z. If the individual isotopes be considered the spread in N/Z is often much greater. This may be seen in Fig. 2, which is sufficiently similar to the diagram in question to illustrate this point. Restriction to atomic numbers greater than about 10 largely eliminates the worst of these discrepancies. Within this range, the larger deviations from the representative line seldom exceed about 15 percent in N/Z among the lower atomic numbers and decrease to half this value in the heavy elements. In elements of odd atomic number the deviations are smaller. A great majority of the points for all the more abundant species fall fairly close to the line. These facts are interpreted to mean that although the form of the equations cannot be closely correct, in a purely numerical sense no serious inaccuracy is represented in the assumptions of (1), (2) and (3). Additional tests, introducing the magnitudes of the constants, are therefore in order.

#### NUMERICAL EVALUATION OF CONSTANTS

From the relations between the constants of (4) and (5) it is found that

$$K_1 = (C_1/C_2 - c_1/c_2)e^2/2r_0 + E_{\alpha}^{\circ}/4 - E_N^{\circ}, \quad (11)$$

$$K_2 = (1/c_2 - 1/C_2)e^2/2r_0, \tag{12}$$

$$k_1 = -(C_1/C_2 + c_1/c_2)e^2/2r_0 + E_{\alpha}^{\circ}/4, \qquad (13)$$

$$k_2 = (1/c_2 + 1/C_2)e^2/2r_0. \tag{14}$$

As a preliminary to the determination of the constants C and c in these equations, and from them the constants K and k, it may be pointed out that Eqs. (4) and (5) are linear in N/Z and  $Z/M^{1/3}$ . Making use of this simplifying feature, N/Z is plotted in Fig. 2 against  $Z/M^{1/3}$  for the isotopes of all elements that have been analyzed by the mass-spectrograph, and for the radioelements. Where more than one isotope is known

<sup>&</sup>lt;sup>3</sup> Harkins, Phys. Rev. 37, 1180 (1931).

<sup>&</sup>lt;sup>4</sup> The isotopic number invented by Harkins represents in terms of the present theory simply the excess, N-Z, of <sup>5</sup> Latimer, J. Am. Chem. Soc. 53, 981 (1931). See also

Latimer and Libby, J. Chem. Phys. 1, 133 (1933).



FIG. 2. Stability boundaries for alpha- and beta-processes.

for a given element, points connected by a line are shown for the heaviest and lightest species. For the radioactive elements all isotopes are shown. They are represented by open circles, with strokes pointing downward to designate emitters of beta-particles and upward for the alpha-emitters.<sup>6</sup>

The lines marked 1 beta and 1 alpha in the figure represent the condition adopted by Heisenberg for zero energy of emission of the beta- and alpha-particles. These curves, however, fail to meet the quantitative tests described later. Numerous variations of them, with the obvious faults corrected but still based on the idea that they should skirt, as far as possible, the edges of the field of points, were therefore tested. None of these proved successful in all respects. This was interpreted to mean that stability alone is not fully effective in determining the boundaries of this field. The existence of unstable species of very long life periods, and fluctuations from the mean positions, largely obscure the proper placement of the curves. Resort was made, therefore, to indirect criteria of correctness in placement.

#### Theoretical conditions affecting boundary curves

The constants in Eqs. (4), (5) and (10) are theoretically related as follows:

$$C_2 = 3D_2/(D_1 + 1), \tag{15}$$

$$c_2 = 3D_2/(D_1 - 1), \tag{16}$$

$$C_2/c_2 = (D_1 - 1)/(D_1 + 1).$$
 (17)

The slope, therefore, of either of the stability boundaries desired for Fig. 2, is theoretically determinable by (15) and (16) from the intercept and slope of the line in Fig. 1. And the ratio of these slopes is fixed in terms of the intercept  $D_1$ alone.

In conformity with these equations, attempts were made to place lines of the required slopes in Fig. 2. Those marked 2 beta and 2 alpha in the figure, for example, were chosen so that constants derived from them reproduce as to order of magnitude the observed energies of the beta- and alpha-disintegrations of the heavy elements. Although they meet successfully these requirements and several other tests that may be made, they fail in other respects. It seems impossible to accept such a placement, and no other based on strict application of (15) and (16) has been found satisfactory.

Were we dealing with a rigorous theory, the discrepancies above could only mean a failure of the basic postulates, or incorrectness in interpretation of the nature of one or the other of the radioactive processes. The beta-process is by no means above suspicion in this last respect, and its correct formulation must remain as an important possibility in removal of the disagreement. Since the theory is but roughly approximate in character, however, neither of these alternatives can be claimed. A part, but not the larger part, of the discrepancy is indeed due to the fact that  $D_1$  and  $D_2$  are of the nature of average values, not exactly applicable to points far from the mean positions in the diagrams. The principal difficulty, however, may lie in an extreme sensitivity, often found in similar connections of the conditions imposed by equating the second derivatives. Regardless of what causes the difficulty, it has proved sufficient in all quantitative tests discussed later to treat Eq. (10) as a rough numerical requirement only. In the final placement of the curves, next to be discussed, the demands of (10) are given some weight, since the later energy equations are otherwise rather meaningless, but the attempt at strict fulfillment is abandoned.

 $<sup>^{6}</sup>$  It is assumed that the beta-activity of potassium and rubidium is due to isotopes 41 and 87, respectively, and the alpha-activity of samarium is attributed to Sm<sup>147</sup>.

#### Final choice of stability boundaries for the betaand alpha-changes

Among all the changes that nuclei may undergo, the emission of beta-particles is unique in one respect. Its reversal,<sup>7</sup> in such cases as are energetically favorable, apparently should require the assistance of no external agency to supply the particle. The s electrons of the extranuclear shells are directed toward the nucleus. Even though the de Broglie wave-lengths of these electrons at every point in their fall through the Coulomb field be much different from nuclear dimensions, making the probability of capture small for a single encounter, this may be compensated by the high frequency of approach. A calculation attributed<sup>8</sup> to Gamow, for instance, shows that the K electrons of nitrogen "penetrate" the nucleus about 1020 times per second. The infrequent occurrence of isobars of unit difference in atomic number may perhaps be taken as direct evidence that such reversals can in fact occur. For there is no reason to suppose that the processes of atomic evolution, either of decay or synthesis, will not often produce such isobaric pairs. If relatively rapid beta-changes are possible in both the forward and the reverse direction, only stable members of such pairs should persist. If, however, the reaction is of unusually long period in either direction, both the stable and unstable species should exist. Actually they are seldom found (compare a later section). We may therefore assume tentatively that the reaction of one of the external electrons with the nucleus is of sufficient probability to require consideration.

It then becomes clear that the line of zero energy of the beta-change should be centrally placed with respect to the existing nuclei. Its strict application would indeed require that the nuclei be restricted to a narrow field, with only one stable isotope of each element. This behavior is actually approached by the elements of odd atomic number, which seldom show more than one abundant isotope. If one recognizes the structural influences previously pointed out, or assumes a nearly flat minimum in the curve of nuclear energy as a function of the number of neutrons associated with a fixed number of protons,<sup>9</sup> the opportunity for a somewhat wider spread of the isotopes becomes apparent, particularly in elements of even atomic number. The more or less central location of the betastability curve remains, however, most probable.

The line marked 3 beta in Fig. 5 has therefore been chosen as representing approximately the boundary of beta-stability toward which the elements tend. With this boundary fixed, there are various limitations which apply to the alpha-boundary and which serve as criteria of the correctness of any position assumed for it, or for the two lines together. These criteria may be listed as follows: (1) the approximate equality of the two forms of the second derivative of the nuclear energy, Eqs. (7) and (9); (2) absence of consistent trend in the constant of integration of Eq. (19) below; (3) agreement of required value of  $r_0$  with values obtained by other methods; (4) correctness as to order of magnitude of the energy effects in known alpha- and beta-disintegrations; (5) correctness in magnitude of energy effects in collision processes; (6) agreement of calculated binding energies of neutron and proton with those obtained from atomic masses in light elements.

Of the many combinations that have been tested by these criteria, the 3 beta—3 alpha pair of Fig. 2 is, on the whole, the most satisfactory. The constants of these curves are  $C_1=0.885$ ,  $C_2=0.0478$ ,  $c_1=-1.600$ , and  $c_2=0.2863$ . Appraisal of these values by criteria 2–6 may be made from results in following sections. Concerning the first criterion, it remains only to say that these constants determine the broken line in Fig. 1. Numerically, for average values of N/Z at Z=90, the second derivative according

<sup>&</sup>lt;sup>7</sup> The reverse of the beta-ray change might be formulated as the ejection of a positron. Observation of an artificially induced process of this type is reported by Joliot and Curie, Nature **133**, 201 (1934). In net effect this would be equivalent to the capture of a beta-particle by the nucleus, which is assumed above to constitute the reverse process. Exact equivalence requires, of course, equality in the energy involved in the two processes. The available energy of the change may determine the process by which it occurs. Thus the ejection of the positron must evolve at least a million electron-volts, whereas the introduction of the electron apparently might occur even with a smaller energy change. When the energy is of the required magnitude, positron emission appears the more probable of the two reverse-beta mechanisms.

<sup>&</sup>lt;sup>8</sup> cf. Chadwick, Constable and Pollard, Proc. Roy. Soc. A130, 480 (1931).

<sup>&</sup>lt;sup>9</sup> Heisenberg, reference 1, II, p. 157, 158, explains the existence of isobars with Z even by curves of energy against N, M constant. The curve above is similar in form.



FIG. 3. Definition of nuclear radii.

to (7) is  $3.01 \times 10^{-7}$ , and by (9)  $2.22 \times 10^{-7}$ . At Z=10 the corresponding figures are  $18.2 \times 10^{-7}$ and  $18.1 \times 10^{-7}$ , with energy in ergs.

#### Magnitude of the proportionality constant $r_0$ , and definition of nuclear radii

The constant  $r_0$  of Eq. (3) is necessary in the solution of Eqs. (11-14). It has been treated in this work as an adjustable constant, chosen simultaneously with the constants C and c to secure the best apparent agreement according to the previously listed criteria. This amounts to a determination of this constant from the other data employed. The value found is 1.6  $\times 10^{-13}$  cm.

This value is in agreement with those obtained by other methods. Gamow,10 from the quantum-mechanical relation between the energy changes and decay constants, calculated the "effective radius" of the nucleus of each of the principal alpha-ray emitters. From his values of  $R_0$ ,  $r_0$  is found to vary from about 1.28 to  $1.40 \times 10^{-13}$  for these elements. Rutherford, Chadwick and Ellis<sup>11</sup> conclude, from studies by Bieler and by Rutherford and Chadwick of the deviations from Coulomb's law in wide angle scattering of alpha-particles by the nuclei of aluminum and magnesium, that  $r_0$  may be taken as about  $1.2 \times 10^{-13}$ . The definition of  $R_0$  is not quite the same in the two last methods of estimation, and in neither of them is it identical with that implied in the present calculation. The relations between the various definitions are made clear in Fig. 3.

In this diagram, the lower curve is of the type usually adopted for the representation of the potential energy, U, of a system consisting of a nucleus and a positive particle as a function of the distance, R, between their centers. The curve is in this case drawn to scale, down to U=0, from the equation  $U=2Ze^2/R-a/R^3$ , with the constant a as determined by Bieler for the alphaparticle in the field of the aluminum nucleus. The upper curve is the corresponding Coulombic function.  $U_0$  represents the lowest energy level,  $U_1$  and  $U_2$  excited levels.  $U_M$  is the maximum potential that the system can possess according to Bieler's treatment.  $R_M$ ,  $3.44 \times 10^{-13}$  cm, is the radius resulting from the scattering experiments.  $R_0$ , the distance at which the purely Coulombic potential is equal to  $U_M$ , is the radius of the nucleus defined by the present calculations. (Based on Bieler's determination of  $U_M$ , it has the value  $5.15 \times 10^{-13}$  cm as compared with  $4.8 \times 10^{-13}$  determined, entirely independently of the theoretical relation between the two definitions by the present calculations.)  $R_2$  results from the quantum-mechanical treatment as applied in Gamow's theory to the level  $U_2$ . As the degree of excitation varies, the radius thus calculated also changes. It approaches  $R_0$ as a limit at high excitation,<sup>12</sup> and at lower levels may be equal to  $R_M$ , as it is for  $U_1$ , or be less than this value. The radius determined by the present calculations is, therefore, theoretically somewhat larger than the others, as was found, and allowing for this, agrees with them excellently.

The present method of determination involves a great variety of data and appears to yield a pretty reliable estimate of  $r_0$ . The definition of this constant in the present theory has also the advantage that its use in Coulomb's law gives

<sup>&</sup>lt;sup>10</sup> Gamow, Der Bau des Atomkerns und die Radioaktivität,

S. Hirzel, Leipsig, 1932, pp. 58-65. <sup>11</sup> Rutherford, Chadwick and Ellis, *Radiations from Radioactive Substances*, The Macmillan Co., New York, 1930, p. 280.

<sup>&</sup>lt;sup>12</sup> The discovery by Gamow that the effective radius in his calculations is greater for excited than for normal nuclei appears therefore to result in part from the nature of his approximations, and does not necessarily imply any change in  $R_M$ , for example.

directly the height of the potential barrier, which Bieler's theory does by use of an additional constant, and Gamow's does not do at all.

#### The mass of the neutron

To determine  $E_N^{\circ}$ , the mass of the neutron must be known. Chadwick<sup>13</sup> obtained its atomic weight as 1.0067. Various other values have since been proposed. The best value is now uncertain and will no doubt remain subject to modification for some time in the future. For the purposes of the present work Chadwick's figure has been adopted. The corresponding value for the energy of formation is  $E_N^{\circ} = 0.159$  $\times 10^{-5}$  erg per neutron. In many of the calculations of this paper the value chosen for  $E_N^{\circ}$  is without influence on the final result. Wherever this is not the case, comment is made.

#### Other auxiliary constants

The energy of formation of the helium nucleus from protons and electrons is taken as  $E_{\alpha}^{\circ} = 4.285 \times 10^{-5}$  erg per atom.

The electronic charge,  $4.767 \times 10^{-10}$  e.s.u., as given by Birge<sup>14</sup> is used here. Avogadro's number correspondingly is taken as  $6.068 \times 10^{23}$ .

#### Final values of constants of Eqs. (1) and (2), and the range of their validity

On substituting the values just discussed of the various constants appearing in Eqs. (11–14), it is found that, in ergs,

$$K_1 = 2.624 \times 10^{-5}, \qquad k_1 = 0.153 \times 10^{-5}, \ K_2 = -1.237 \times 10^{-5}, \qquad k_2 = 1.733 \times 10^{-5}.$$

Of these constants,  $K_1$  alone is dependent on the mass of the neutron.

The intention of Heisenberg was to restrict his theory to the heavy elements only. In this work, however, no very urgent reason for rigid restriction of this kind has appeared. In Figs. 1 and 2, for example, the fluctuations, periodic and otherwise, increase only gradually in magnitude from the heavy to the light elements. They become exceptionally large, however, below atomic number 10 and here, also, the packing fractions of different isotopes lie on divergent curves. Up to this value of Z, numerical approximations of the theory (such as the equality of Z and Z-1 or Z-2, and the equivalence of integrals with the corresponding summations) may also introduce appreciable errors. For these reasons the application of the above constants has been restricted to elements with Z > 10.

#### NUCLEAR ENERGIES

The energy of formation of any nucleus from neutrons and protons is approximately expressible in terms of N and Z and the constants above. This quantity will be represented by E and taken as  $-\Delta E$  for the process of synthesis. From Eqs. (6) and (8),

$$dE = (K_1 + K_2 N/Z) dN + (k_1 + k_2 N/Z - e^2 Z/r_0 M^{1/3}) dZ.$$
(18)

Integration of this equation, assuming N/Z to be maintained constant during the synthesis of the nucleus, yields

$$E = (K_1 + k_2)N + K_2N^2/Z + k_1Z - 3e^2Z^2/5r_0M^{1/3} + I. \quad (19)$$

Here I is the constant of integration.

With values of E determined from the masses<sup>15</sup> of all nuclear species of Z > 10 for which a measured value of the packing fraction has been published, adjustment of the constants was made to secure constancy in I and to meet simultaneously the criteria 4 and 5 above. The results are shown in Table I.

The degree of constancy of I may be seen in the last two columns. These include, respectively, the algebraic deviation of individual values of I from the mean, -9.6, of all, and the experimental uncertainty resulting from the error in the determinations of isotopic weight.<sup>16</sup>

<sup>&</sup>lt;sup>13</sup> Chadwick, Proc. Roy. Soc., A136, 692 (1932).

<sup>&</sup>lt;sup>14</sup> Birge, Phys. Rev. 42, 736 (1932).

<sup>&</sup>lt;sup>15</sup> Now conveniently collected in Aston's recent book, Mass Spectra and Isotopes, Longman's, Green and Co., New York, 1933.

<sup>&</sup>lt;sup>16</sup> The experimental uncertainties given in Table I are based on the author's estimates of error in the original papers, and are usually "limits." In some instances these have since been replaced by "probable errors" in Aston's tabulation. They are listed in the table opposite the isotope for which the primary determination of mass was made, and with which the other isotopes given were compared. In certain instances a numerical estimate of error was not originally given by Aston, the measurement being designated as "rough" or "provisional." In such cases, the experimental uncertainty is given in the last column of the table as greater than that estimated for neighboring elements. Aston has not published a definite packing faction for lead, but makes the statement that the atomic weights of the isotopes studied are integral with those of mercury to 1 or 2 parts in 10,000.

The mean deviation from the average I is 1.5, and the average experimental uncertainty is 2.8. Of the elements in the table from  ${}_{24}Cr^{58}$  to  ${}_{82}Pb^{208}$ , there is none for which the deviation from the average I significantly exceeds the experimental uncertainty.

Eq. (19) is of some intrinsic interest as a fairly successful attempt at an energy equation. Over much of its range of application, nuclear energies or isotopic weights can be calculated with as much accuracy (within about 10 million electronvolts or 0.01 atomic weight unit on the average) as they have hitherto been measured. As more accurate experimental values for the heavy elements become available in the future, this will no doubt no longer be possible by so simple an equation. Moreover, energy relations in particular nuclear processes can usually be more satisfactorily treated by equations specifically applicable, of types discussed later. The principal interest of the general energy equation centers therefore, in the present connection, in its bearing upon the self-consistency and numerical reliability of the constants involved.

The values of the energies of formation in Table I are greatly affected by the mass adopted for the neutron. Fortunately, the integration constant, I, is independent of  $E_N^{\circ}$ . The desired tests are therefore uninfluenced by uncertainties in it.

#### ENERGY OF EMISSION OR CAPTURE OF PARTICLES BY NUCLEI

The change in energy that accompanies an emission or (with reversed signs) a capture of any particle of zero kinetic energy is conveniently formulated as  $\Delta E = \overline{E} - V$ , where Vrepresents the Coulombic energy. For the neutron and proton, the binding energies are, of course, given by Eqs. (1) and (2). The binding energy of any composite particle is obtainable in terms of the simple ones. For the alphaparticle,

$$\overline{E}_{\alpha} = 2(K_1 + k_1) - E_{\alpha}^{\circ} + 2E_N^{\circ} + 2(K_2 + k_2)N/Z. \quad (20)$$

Inserting numerical values of the constants,<sup>17</sup>

$$\overline{E}_{\alpha} = 1.587 + 0.992 N/Z.$$
 (20a)



FIG. 4. Energy of alpha-particles as function of N/Z of parent atom in series of isotopes.

Similarly, for the deuton,

$$\overline{E}_{D} = K_{1} + k_{1} + E_{N}^{\circ} - E_{D}^{\circ} + (K_{2} + k_{2})N/Z, \quad (21)$$

$$\overline{E}_D = 2.652 + 0.496 N/Z.$$
 (21a)

#### Energy of emission of alpha-particles

If the kinetic energy of the emitted alphaparticle is denoted by  $W_{\alpha}$ , we may write approximately,  $W_{\alpha} = V_{\alpha} - \overline{E}_{\alpha}$ . This becomes

$$W_{\alpha} = E_{\alpha}^{\circ} - 2E_{N}^{\circ} - 2(K_{1} + k_{1}) - 2(K_{2} + k_{2})N/Z + 2e^{2}Z/r_{0}M^{1/3} \quad (22)$$
or

$$W_{\alpha} = -1.588 - 0.992 N/Z + 0.284 Z/M^{1/3}$$
, (22a)

The general requirements of Eq. (22) in relation to the experimental facts are clearly brought out by the following considerations. The factor  $M^{1/3}$  in the last term of (22) varies less than 2 percent among the isotopes of any radioactive element. For any constant value of Z, therefore, the energy of the emitted alpha-particle should, according to (22), be a nearly linear function of N/Z. This is tested graphically in Fig. 4. The points of this figure represent the observed energies of the particles plotted against N/Z. Those belonging to the isotopes of a single element are connected by lines marked with the

 $<sup>^{17}</sup>$  In all numerical equations the resultant energies are in units of  $10^{-5}$  erg per atom. In the tables and tests, comparisons are sometimes made in other units. Conversion factors are  $10^{-5}$  erg =  $6.28 \times 10^{6}$  electron-volts, and  $10^{-5}$  erg per atom = 0.00675 g per g atom,  $O^{16}\!=\!16$ .

	-Element	;	Ė. (	τ.		Exp't'l	-	-Element		E f	T		Exp't'l
Z	Symbol	М	formation	Eq. 19	$(I - I_{Av.})$	tainty	Z	Symbol	М	formation	Eq. 19	$(I - I_{Av.})$	tainty
10	Ne	20 22	21.9	-7.7	1.9	0.1	52	Te	126	142.6	-11.5 -11.4	-1.9 -1.8	4.0
14	Si	28	32.6	- 7.8	1.8	0.4	53	I	127	144.5	-10.5	-0.9	3.7
17	Cl	35	40.0	-10.0	-0.4	0.7	54	Ас	124	143.5	-10.0	-0.2	
18	А	36	42.4	-10.9 -8.4	1.2	0.6	-		120	145.0	-10.1 -10.3	-0.3 -0.7	
24	Cr	40 52	40.8 64.1	-10.4 -8.0	-0.8	2.2			130	147.8	-10.3 -10.2	-0.6	
28 30	Zn	58 64	70.6 77.6	-7.3 -8.2	2.3 1.4	(2.2?) >2.5?			132 134	150.0	- 9.7 - 9.5	-0.1	4.0
33 34	As Se	75 78	89.2 91.7	-10.6 -11.6	-1.0 -2.0	1.6 > 2.4	55	Cs	136 133	$154.3 \\ 150.5$	-8.8 -10.2	-0.8	4.0
35	Br	80 79	93.5 94.5	-12.3 - 9.8	-2.7 -0.2	$\begin{array}{c} 2.4 \\ 1.8 \end{array}$	56 73	Ba Ta	$\frac{138}{181}$	158.3 201.8	-7.1 -1.9	$2.5 \\ 7.7$	4.0 > 5.5?
36	Kr	81 78	96.9 94.1	-10.0 - 7.9	-0.4 1.7	2.4	75 76	Re Os	187 190	$200.2 \\ 203.4$	- 8.4 - 7.7	1.2 1.9	5.5 5.6
		80 82	96.1 97.9	-9.0 -10.1	-0.6	$\begin{array}{c} 2.4 \\ 1.8 \end{array}$	80	$_{\rm Hg}$	192 196	$\begin{array}{c} 205.4 \\ 204.8 \end{array}$	-6.8 -10.6	2.8 - 1.0	
		83 84	98.9 99.8	-10.3 -10.5	$-0.7 \\ -0.9$	$\begin{array}{c} 1.8\\ 1.9 \end{array}$			197 198	$\begin{array}{c} 205.8\\ 206.8\end{array}$	-10.6 -10.6	-1.0 -1.0	5.9
41	Сь	86 93	$101.6 \\ 109.8$		-1.3 -0.3	1.9 > 2.8?			199 200	$207.7 \\ 208.7$	-10.6 -10.3	$-1.0 \\ -0.7$	
42	Mo	98 100	$111.9 \\ 114.0$	-13.5 -13.7	-3.9 -4.1	>3.0?			201 202	209.7 210.7	-10.2 -10.1	-0.6 -0.5	
50	Sn	112 114	$131.2 \\ 133.3$	-7.4 -80	2.2				203 204	211.7 212.7	-9.7 -9.5	-0.1	
		115	134.5 135.7	- 8.0	1.6		81	Tl	203	204.9	-16.3	-6.7	5.9
		117	136.6	- 8.3	1.3		82	РЬ	200	215.0	- 8.8	0.8	(5.9?)
		110	137.7	-8.3	1.1	26			207	217.0	- 8.5	1.1	
		120	140.1	- 8.2	1.3 1.4	5.0				Average	- 9.56	1.5	2.8
		124	144.4	- 7.6	2.0								

TABLE I. Energy of formation of nuclei from neutrons and protons. (The unit<sup>17</sup> of energy in each of the last four columns is  $10^{-5}$  erg.)

atomic number to which they correspond. With the exception of two points belonging to element 84, the required variation of energy with N/Z is roughly substantiated. The essentials of this relation were first discovered and discussed by Fajans<sup>18</sup> who pointed out a connection between atomic weight and stability.

A second requirement of Eq. (22) is, however, contradicted in Fig. 4. At constant values of N/Z the expectation is that the energy of the alpha-particle should be greater the larger the value of Z. This is not fulfilled, and presents an interesting problem. It is related, obviously, to the fact that in a series of successive alphadisintegrations the energy of the particles, as a

rule, progressively increases. This can be taken as an indication that structural effects, neglected in the simple theory, are of some importance here. It is possible that the removal of particles from a completed group would leave the remaining structure progressively less stable until the next sub-group is approached. If this is not the explanation, the behavior is still explicable on the basis of the failure of the tacit assumption that excited states are not involved. If the departure of an alpha-particle, for example, leaves the residual nucleus with an excitation which, because of some sort of forbidden transition, cannot be relieved by radiation, the particles from the residual nucleus will have greater energies than expected.

Whatever the cause of the difficulty, it is apparent that faithfulness to detail will not be

<sup>&</sup>lt;sup>18</sup> Fajans, Le Radium 10, 171 (1913); Naturwiss. 14, 963 (1926): *the Baker Lectures*, Cornell University, McGraw Hill Book Co., N. Y. 1931.

Element	Isotope	$W_{\alpha}$ (obs.)	$W_{\alpha}$ (calc.)	Differ- ence
83 Ac C	211	1.058	0.842	0.216
Th C	212	0.962	0.824	0.138
Ra C	214	0.876	0.786	0.090
84 Ra F	210	0.840	0.936	-0.096
Ac C'	211	1.200	0.918	0.282
Th C'	212	1.402	0.898	0.504
Ra C'	214	1.228	0.866	0.362
Ac A	215	1.184	0.846	0.338
Th A	216	1.078	0.830	0.248
Ra A	218	0.956	0.794	0.162
86 An	219	1.092	0.930	0.162
Tn	220	1.000	0.912	0.088
Rn	222	0.870	0.874	-0.004
88 Ac X	223	0.900	1.010	-0.110
Th X	224	0.902	0.994	-0.092
Ra	226	0.755	0.960	-0.205
90 Ra Ac	227	0.942	1.090	-0.148
Ra Th	228	0.850	1.076	-0.226
Io	230	0.726	1.040	0.314
Th	232	0.676	1.006	0.330
91 Pa	231	0.797	1.098	-0.301
92 U H	234	0.740	1.122	-0.382
ŬÎ	238	0.648	1.042	-0.384
		1. 1 2.	Average	0.26

TABLE II. Energy of alpha-particles. (The unit<sup>17</sup> of energy is  $10^{-5}$  erg per particle.)

	Element	Isotope	Obs. energy of par- ticles (Aver- age)	Obs. energy of photons (Average per particle)	Total energy (Obs.)	Calcu- lated energy
•	81 Th C"	208	0.70			0.52
	82 Ra D Th B Ra B	210 212 214	0.01 0.10 0.23	0.05 <0.21 0.20	$0.06 < 0.31 \\ 0.43$	0.29 0.79 1.3
	83 Ra E Th C Ra C	210 212 214	0.34 0.7 0.76	0.0 1.66	$\frac{0.34}{2.42}$	-0.43 0.05 0.53
	90 UX I	234	0.11	·		0.31
	91 UX II	234	0.39			-0.40
	19 K	41?	0.4	0.06	0.46	0.15
	37 Rb	87?	0.15		0.15	1.2

TABLE III. Energy of beta-ray changes. (The unit<sup>17</sup> of energy

is 10<sup>6</sup> electron-volts.)

achieved by Eq. (22). Average behavior of the radioactive group as a whole must, however, be reflected in the equation if the constants are correctly chosen. Adjustment of them has been made with this in mind.

In Table II the observed and calculated energies are compared for the alpha-emitters appearing in Fig. 4. The average deviation of the calculated from the observed energies corresponds to  $1.6 \times 10^6$  electron-volts, or about 0.0015 atomic weight unit. Although this is large compared to the error of direct measurement, it represents an accuracy about 20 times that of the average at present achieved above mass number 100 by the use of the mass-spectrograph. For calculations relating to nuclear species which, because of very long life period or very short range of particle, cannot be directly studied, a considerable increase in accuracy over the only other method of calculation available is therefore gained in this range. This, and the approximate substantiation of the constants employed, constitute the only value of (22), which obviously does not contain all of the factors requisite for close approximation.

#### Energy of beta-ray changes

Measurements of the mean energy of betaparticles are available for nine of the radioelements.<sup>19</sup>. For five of these the mean energy, per disintegration, of the photons emitted may be inferred from data cited by Rutherford, Chadwick and Ellis.<sup>20</sup> These data are compared in Table III with energies calculated by the equation

$$W_{\beta} = k_1 - K_1 - E_N^{\circ} + (k_2 - K_2)N/Z - e^2 Z/r_0 M^{1/3}, \quad (23)$$

which, with the present constants, becomes

$$W_{\beta} = -2.630 + 2.970 N/Z - 0.142 Z/M^{1/3}$$
. (23a)

The energies calculated by (23) presumably represent the total energy of the change. For the cases shown in the table the average difference of the calculated from the observed total energies is less than a million volts. The agreement is, therefore, as good as in any other part of this work. The curve 3 beta was placed in Fig. 2 so as to fall between the lowest isotopes of the even and odd numbered beta emitters among the heavy radio-elements. This is reflected in Table

<sup>&</sup>lt;sup>19</sup> See Gamow, reference 10, p. 67 for detailed references. <sup>20</sup> Reference 11, pp. 401, 403, 501.

Particle captured	Original and final nuclei	Observed decrease in mass	Calcu- lated decrease in mass	Differ- ence calcobs.	Exp't'l uncer- tainty	Particle captured	Original and final nuclei	Observed decrease in mass	Calcu- lated decrease in mass	Differ- ence calcobs.	Exp't'l uncer- tainty
Proton	$\begin{array}{r} Be^9 & -B^{10} \\ B^{11} & -C^{12} \\ C^{13} & -N^{14} \\ N^{15} & -O^{16} \\ O^{18} & -F^{19} \\ F^{19} & -Ne^{20} \end{array}$	0.0098 .0152 .0038 .0110 .0143 .0110	0.0138 .0129 .0122 .0117 .0127 .0108 Average	$\begin{array}{r} 0.0040 \\0023 \\ .0084 \\ .0007 \\0016 \\0002 \\ .0029 \end{array}$	0.002 .003 .006 .003 .005 .003 .0037	Neutron	$\begin{array}{c} {\rm Li}^6 & -{\rm Li}^7 \\ {\rm B}^{10} & -{\rm B}^{11} \\ {\rm C}^{12} & -{\rm C}^{13} \\ {\rm N}^{14} & -{\rm N}^{15} \\ {\rm O}^{16} & -{\rm O}^{18} \\ {\rm Ne}^{20} - {\rm Ne}^{22} \end{array}$	0.0066 .0092 .0063 .0115 0008 .0093	0.0094 .0094 .0094 .0094 .0115 .0118 Average	$\begin{array}{c} 0.0028\\.0002\\.0031\\0021\\.0107\\.0025\\e^{21}\\.0037\end{array}$	0.001 .003 .004 .006 .003 .002 .0036
Deuton	$\begin{array}{ccc} Li^7 & -Be^9 \\ Be^9 & -B^{11} \\ B^{10} & -C^{12} \\ B^{11} & -C^{13} \\ C^{12} & -N^{14} \\ C^{13} & -N^{15} \\ N^{14} & -O^{16} \\ O^{18} & -Ne^{20} \end{array}$	0.0127 .0181 .0235 .0206 .0092 .0144 .0216 .0234	0.0197 .0199 .0190 .0198 .0187 .0182 .0185 .0192 Average	$\begin{array}{c} 0.0070\\ .0018\\0045\\0008\\ .0095\\ .0038\\0031\\0042\\ 2^{21}0.0041\end{array}$	.001 .002 .003 .004 .004 .006 .003 .004 .0037	Alpha- Particle	$\begin{array}{cccc} Li^6 & -B^{10} \\ Li^7 & -B^{11} \\ Be^9 & -C^{13} \\ B^{10} & -N^{14} \\ B^{11} & -N^{15} \\ C^{12} & -O^{16} \\ N^{15} & -F^{19} \\ O^{16} & -Ne^{20} \\ O^{18} & -Ne^{22} \end{array}$	$\begin{array}{c} 0.0032\\.0058\\.0137\\.0077\\.0100\\.0058\\.0054\\.0055\\.0140\end{array}$	0.0142 .0166 .0154 .0130 .0144 .0124 .0129 .0113 .0132 Average	$\begin{array}{c} 0.0110\\ .0108\\ .0017\\ .0053\\ .0044\\ .0066\\ .0075\\ .0058\\0008\\ e^{21}0.0046\end{array}$	$\begin{array}{c} 0.002\\.002\\.004\\.004\\.004\\.001\\.005\\.001\\.004\\.0033\end{array}$

TABLE IV. Decrease in mass on capture of particles by nuclei of light elements. (The unit<sup>17</sup> is the gram per gram atom,  $O^{16}=16.$ )

III by the fact that the calculated energies are usually too high for the even numbered and too low for the odd numbered elements. Better agreement would be obtained by treating these classes separately. This has been sacrificed throughout the present work for the sake of representing average behavior and general trends.

### Energy evolved in capture of various particles by nuclei of light elements

A check of the constants derived here when applied to nuclei below the heaviest elements is to be desired. Unfortunately the only available data of sufficient experimental accuracy to permit reliable tests pertain to the elements below atomic number 10. In order to make sure that the proposed constants do not lead to absurd results even in this region, calculations have been made of the energy which would be liberated if a proton, a neutron, a deuton or an alphaparticle were captured by various light nuclei. The calculated energies are compared in Table IV with the decrease in mass corresponding to the change.

The calculated values will be referred to as  $\Delta E$  and are based on the following equations:

$$\Delta E_P = -0.153 - 1.733 N/Z + 0.142 Z/M^{1/3}; \quad (24)$$

$$\Delta E_N = -2.624 + 1.237 N/Z; \tag{25}$$

$$\Delta E_D = -2.652 - 0.496 N/Z + 0.142 Z/M^{1/3}; \quad (26)$$

$$\Delta E_{\alpha} = -1.587 - 0.992N/Z + 0.284Z/M^{1/3}.$$
 (27)

In applying Eqs. (24)–(27) to the processes indicated in Table IV, the values of N and Zare taken to correspond to the nucleus to which the particle is added. The calculated values of  $\Delta E_N$  are greatly affected by the mass adopted for the neutron, but the "observed" values are also affected in the same way. The difference between the observed and calculated results is therefore independent of the mass of the neutron.

The average difference of the calculated from the observed results in Table IV varies between about 2.7 and 4.3 million volts for the different processes while the average experimental uncertainty is of the order of 3.5 million volts.<sup>21</sup>

As applied to the very light elements in Table IV, the interest in Eqs. (24)–(27) centers entirely in the control afforded of the constants selected. In this region the basic curves adopted as standard seem to introduce definite, but not unduly large, trends away from the experimental values in the case of the alpha-particle. However, the trends and individual fluctuations should

 $<sup>^{21}</sup>$  These differences are computed omitting the figures for lithium, on the basis of its extreme lightness, from the averages. The experimental uncertainty in the last column is based on an estimate of 0.003 unit for C<sup>13</sup>, N<sup>15</sup> and O<sup>18</sup>, with the observers estimates employed for the remaining elements.



FIG. 5. Binding energies.

decrease with approach to the region in which the original assumptions are likely to be more closely valid. It is believed therefore that the equations afford reliable approximations, of greater accuracy than now obtainable from isotopic masses, for nuclei of large mass number.

#### NUCLEAR REACTIONS IN COLLISIONS WITH PARTICLES

## General characteristics deducible from binding energies

Certain deductions of rough qualitative character concerning collision processes are obtainable from consideration of the binding energies alone of the various particles. This is facilitated by Fig. 5, where graphs<sup>22</sup> are shown of binding energies as functions of N/Z, based on Eqs. (1), (2), (20) and (21).

One of the features of the diagram is the large magnitude at all values of N/Z of the binding energy of the deuton. This implies relatively marked effectiveness of this particle in ejection of others.

According to the diagram, the alpha-particle should always be more effective, from a purely energetic standpoint, than the proton in producing disintegrations, though for equal kinetic energy of the particles the proton has a higher probability of surmounting or penetrating the Coulomb barrier. The two curves approach each other at large values of N/Z. There is also some indication from Table IV that the alpha-curve should be lowered somewhat relative to that of the proton among light elements.

Neutrons of low kinetic energy will, according to Fig. 5, be incapable of liberating other particles in their interactions with nuclei. They should be ejected readily by any of the other particles, especially if the struck nucleus is of high neutron-proton ratio, as it is, relative to neighboring elements, in Li<sup>7</sup>, Be<sup>9</sup> and B<sup>11</sup> for example.

The question of capture or escape of any projectile particle which has caused a disintegration is also answered, in part, by Fig. 5. Excluding the neutron, the smallest of the binding energies is of the order of 12 million volts. Neither natural particles nor artificially accelerated particles of such high energy have yet been employed in nuclear experiments. Disintegration without capture is therefore improbable or impossible in most cases. The ejection of a neutron without capture of the colliding particle is, on the other hand, indicated to be quite possible in many cases as far as total energy, independent of questions of transfer, is concerned.

The above qualitative conclusions seem to be borne out by experience. They are of some value for orienting purposes, but many other factors enter any more detailed consideration. Adequate quantitative treatment from the energetic standpoint of any nuclear collision requires a complete formulation of the process. Given such a formulation, equations applicable to the process can readily be deduced, as illustrated below.

#### Energy of particles ejected in nuclear disintegration by collision

With symbols previously employed and subscripts 1 and 2 to denote the projectile and emitted particles, respectively, the energy of the ejected particle is

$$W_2 = W_1 - V_1 + \overline{E}_1 - \overline{E}_2 + V_2. \tag{28}$$

This equation assumes the capture of the impinging particle, neglects the kinetic energy and

<sup>&</sup>lt;sup>22</sup> The lines of this figure represent, of course, mean positions about which the points of the system fluctuate.

excitation of the residual nucleus, and assumes that only one particle and no photon is ejected in the process. Strictly, the quantities  $V_1$  and  $\overline{E}_1$ are taken to correspond to the original struck nucleus, and  $\overline{E}_2$  and  $V_2$  to the residual nucleus. If, however, the nucleus is sufficiently large, the differences between the original and residual nucleus may be neglected without significant error. This has been done in the present calculations.

Substitution in (28) of the expressions previously obtained ((1), (2), (20) and (21)) for the appropriate binding energies leads to equations covering any type of capture-ejection process. A series of such equations for processes of current interest are recorded below.

 $W_P = W_{\alpha} + 1.434 - 0.741 N/Z - 0.142 Z/M^{1/3}$ , (29)

$$W_P = W_D + 2.499 - 1.237 N/Z, \tag{30}$$

 $W_{\alpha} = W_P - 1.434 + 0.741 N/Z + 0.142 Z/M^{1/3}$ , (31)

$$W_{\alpha} = W_D + 1.065 - 0.496 N/Z + 0.142 Z/M^{1/3}, (32)$$

$$W_N = W_{\alpha} - 1.037 + 2.229 N/Z - 0.284 Z/M^{1/3}$$
, (33)

 $W_N = W_P - 2.471 + 2.970 N/Z - 0.142 Z/M^{1/3}$ . (34)

These equations do not depend in any way upon the mechanism of the assumed processes (penetration or surmounting of barriers, resonance, etc.), and they of course can yield no information as to the probability of occurrence of the various reactions.

The principal data suited to the tests of these equations are the observations of Rutherford and Chadwick<sup>23</sup> on protons ejected by alphaparticles. These are compared in Table V with the calculated values. In addition to these there are fragmentary data relating to other reactions. Lewis, Livingston and Lawrence<sup>24</sup> report the ranges of alpha-particles ejected from magnesium and aluminum by deutons, and Lawrence and Livingston<sup>25</sup> found long range protons ejected from aluminum by deutons. Only elements above atomic number 10 are included in the table.

 TABLE V. Energy of ejected particles in collision processes.

 (Energies<sup>17</sup> are in 10<sup>6</sup> electron-volts.)

Projec- tile particle	Projec- tile energy	Struck nucleus	Ejected particle	Equa- tion applied	Ener Calc.	rgy of ej particle exp't'l	ected Diff.
Alpha Alpha Alpha Alpha Alpha Deuton Deuton Deuton Deuton	7.68 7.68 7.68 7.68 7.68 2.7 1.2 1.2 1.2	Na <sup>23</sup> Mg <sup>24</sup> Mg <sup>26</sup> Al <sup>27</sup> P <sup>31</sup> Al <sup>27</sup> Mg <sup>24</sup> Mg <sup>26</sup> Al <sup>27</sup>	Proton Proton Proton Proton Proton Alpha Alpha Alpha	(29) (29) (29) (29) (29) (30) (32) (32) (32)	8.2 8.3 7.6 7.8 7.5 10. 8.5 7.9 8.4	6.9 5.6 5.6 8.8 7.7 7.5 7.0 7.0 7.0 7.0	$\begin{array}{c} 1.3\\ 2.7\\ 2.0\\ 1.0\\ 0.2\\ 2.5\\ 1.5\\ 0.9\\ 1.4 \end{array}$

#### Threshold energies of projectiles and critical characteristics of the struck nuclei in disintegrations by collision

For ejected particles of any arbitrarily fixed energy, calculation of the threshold energy of the projectile (or critical composition of the struck nucleus) can be made by Eq. (28), independently of any assumption as to mechanism. As the probability of occurrence of a process depends greatly upon this factor it is, however, often desirable to consider it. In illustration, suppose that a process of capture of a particle with ejection of a single particle of another sort is in question. Assume that the external Coulombic barrier can be penetrated frequently by the projectile particle if the total energy effect is favorable to its capture, and that for the ejection of the second particle a quantity of energy essentially equal to its binding energy must be supplied. Simple considerations then lead to the following modification of (28);

$$W_1 - V_1 + E_1 = \overline{E}_2.$$
 (35)

Applying (35) to the ejection of protons on capture of alpha-particles there results

$$N/Z = (1.515 + W_{\alpha})/0.741 - 0.383Z/M^{1/3},$$
 (36)

where N, Z and M pertain (with the approximation introduced in a similar case previously), to the struck nucleus. If these variables are given in magnitude, the value of the critical energy of the alpha-particle may be solved for. If the energy of the alpha-particle is fixed, the critical characteristics of the struck nucleus can be determined. This is most conveniently done by calculating from (36) values of  $Z/M^{1/3}$  corresponding to two arbitrarily chosen values of N/Z. On placing the resulting values on a plot like that in Fig. 2, it will be found that the line

<sup>23</sup> Reference 11, p. 293. The "observed energies" in Table V correspond to the maximum range of the protons in the forward direction.

<sup>24</sup> Lewis, Livingston and Lawrence, Phys. Rev. 44, 55 (1933).

<sup>&</sup>lt;sup>25</sup> Lawrence and Livingston, Phys. Rev. 45, 220 (1934).

determined by them separates the nuclei for which the assumed reaction is indicated to be possible from those for which it is not. On carrying out this process for the alpha-particle of Ra C' (of  $1.222 \times 10^{-5}$  erg energy) it is found that all isotopes of elements below <sub>25</sub>Mn meet the condition. This necessarily rough result may be compared with the experimental one of Rutherford and Chadwick, who found disintegrations of this type as high as <sub>19</sub>K, but not in heavier elements.

The possibilities predicted by Eq. (35) applied to capture of a deuton with ejection of a neutron or a proton are interesting. So far as total energy is concerned, neutrons might be ejected in this way from every element in the periodic system by million volt deutons. With the same assumption as to mechanism as employed above, Eq. (35) predicts that proton ejection should cease, again for million volt deutons, in the neighborhood of 33As. This is the limit at which sufficient energy is supplied on capture to carry a proton completely over the internal barrier. If this condition is abandoned as a necessary feature of the process, emission may occur even among very heavy elements. For example, if a deuton is captured by  $_{78}$ Pt<sup>195</sup>, there is  $1.645 \times 10^{-5}$  erg available for excitation of a proton in the nucleus. Of this amount,  $0.802 \times 10^{-5}$ erg  $=5.04\times10^6$  e.v., represents the excitation above the zero of energy. A nucleus excited to this extent should have a pretty short life and might eject a proton immediately after capture of the deuton.

#### STABILITY RELATIONSHIPS

#### Conditions for spontaneous emission of protons, deutons and neutrons

The question often arises as to whether protons are ever ejected from the nucleus in any spontaneous disintegration. The answer given by the present theory is obtained by making the energy,  $\Delta E_P$ , zero in Eq. (24). The resulting equation in N/Z as a function of  $Z/M^{1/3}$  represents the borderline of stability in such processes. The graph of this line is shown in Fig. 6, together with the similar curves<sup>22</sup> for other processes, including the beta- and alpha-curves determined previously. Points to the left of the



FIG. 6. Stability boundaries for emission of various particles.

proton line in the diagram represent nuclei that are stable with respect to emission of this particle. All existing species lie in the region between the dotted lines. Radioactivity of the type in question is therefore not expected among them. Experimentally, of course, no such spontaneous proton emission has ever been detected. Measured packing fractions negate the idea also in all cases where decisive accuracy is attained. (Compare Table IV.) Such accuracy is lacking in the heavy elements. The result above is therefore of some interest in this region.

Judging from Eq. (26), no existing nuclear species need ever be remotely suspected of spontaneous emission of deutons. The boundary curve for this process lies far from all others, as shown in Fig. 6. The region to the left is again the stable one. Eq. (25) shows that in the average case N/Z must exceed about 2.13 for instability with respect to the emission of neutrons. Unless Chadwick's mass of the neutron is 3 or 4 million volts too high no known nucleus can spontaneously disintegrate in this way.

#### Factors determining the maximum atomic number and field of existence of nuclei

The point at which the periodic system of the elements "ends" has also been the subject of much speculation. If the discussion is limited to the nuclear species that are stable under conditions existing at present on the earth, the limit for such stability is found to be in the vicinity of atomic number 51 (Sb). The alpha-

and beta-boundaries intersect each other in this region. Applying the present ideas strictly, every element above this point should be unstable with respect to one of these disintegrations and those in the region between the two curves above their intersection should be unstable with respect to both. Actually the fluctuations and periodic variations which are found vitiate this as a rigid deduction. It remains, however, strongly supported so far as the average tendencies are concerned, by the very fair agreement found in the quantitative tests of the hypotheses. The relative abundance of the elements above the intersection of the alpha- and beta-curves, and the likelihood of existence of elements above uranium, are resolved, therefore, into questions of rate of decay, or balance between decay and synthesis.

Judging from Fig. 6, the increasing instability with respect to alpha-emission may be the factor finally terminating the system of elements. The heaviest elements are farther from the indicated stable positions for the alpha than for the other changes. Periodic tendencies against the general trend may displace the breaking off point beyond the position where it might be expected that life periods would in the average case become very short. With the ultimate reversal of an opposed tendency such as this, after the completion of a period, for example, the break from relatively long to extremely short life might be sharp. This may happen above uranium.

The approach and final intersection of the dotted lines of Fig. 6 would also determine and "end" of the system. This approach seems too gradual to account for the breaking off at Z = 92. However, the provisional interpretation given previously of these borders of the field of existing nuclei has been well supported by the results based upon it. This aspect of the situation may be outlined as follows. Stability with respect to the beta-change and its reverse determines a single line along which all nuclei should lie. Very slow rates of change make possible the existence of unstable isotopes. The "spread" of the field of points is then determined by the deviation allowable from the stable composition before the electron transfer becomes rapid. Near the point Z=51 the rate of the alpha-change constitutes an additional factor determining the width of the field. This appears to be of secondary importance, however, until the radio-elements are reached.

#### Spontaneous changes among "non-radioactive" elements

The point Z=51 determined above as the mean position at which stability with respect to the alpha-change ends, is surprisingly low. It is in agreement, however, with a similar conclusion from Gamow's<sup>26</sup> curve of nuclear energies against number of alpha-particles composing the nucleus, which goes through a minimum in this region. The large gap between this point and the lowest alpha emitter, Z = 83, of the radioelements, is considerably reduced by the recent discovery<sup>27</sup> of the alpha-activity of samarium, Z=62. Other alpha-changes, that have hitherto eluded discovery because of very low energies or slow decay, are to be anticipated in this region.

Concerning the beta-change, any species lying much above or much below the beta-curve of Fig. 6, may reasonably be suspected of activity involving loss or gain of an electron by the nucleus. There is, indeed, indirect but strong evidence that such changes are actually occurring in certain elements. The mass-spectrograph reveals a number of isobars. Of the total of 34 such isobaric pairs hitherto reported, only 9 are of unit difference in atomic number, and only a few of these are established beyond reasonable doubt. The relatively infrequent occurrence of this type of couple has been employed in the previous argument. We may, however, take the pair  $_{49}In^{115} - _{50}Sn^{115}$  as a probable example of two atomic systems of the same total composition (in protons and electrons), but of different energies. The conclusion is inevitable that in such a system a spontaneous change, which in effect is the transfer of an electron between the nucleus and the exterior, is in progress in one direction or the other. The speed of the change must be finite, though perhaps immeasurably slow, or of undetectably low energy. Mass determinations sufficiently accurate to decide the direction of this change are lacking. Eq. (23) gives an energy of about  $4 \times 10^5$  e.v. favoring

 <sup>&</sup>lt;sup>26</sup> Reference 10, pp. 19, 23.
 <sup>27</sup> Hevesy and Pahl, Nature 130, 846 (1932); Libby and Latimer, J. Am. Chem. Soc. 55, 433 (1933).

the change from Sn to In, but this is no larger than the error likely in such calculations.

There are 24 isobaric couples reported of two units difference in atomic number. In such cases the energy of the change from one member of the pair to the missing intermediate element may be opposite in sign from that between the two members of the couple.<sup>9</sup> A double transfer would then be required to effect the change. A simultaneous emission or absorption of two electrons would no doubt be of extremely low probability, removing such pairs from the list of possibly detectable changes. If, however, the absence of the intermediate isobar merely indicates that this species is of very short life, the overall change need not be so slow. It may be remarked that for the couple 34Se<sup>80</sup>-36Kr<sup>80</sup>, Aston gives the atomic weights 79.941 and 79.926, respectively. The mass difference is not large compared to the possible uncertainty of measurement, yet seems sufficient to justify the belief that Se<sup>80</sup> is unstable with respect to Kr<sup>80</sup>. Eq. (23) also leads to this result.

#### Conclusion

The very simple postulates of Heisenberg appear to have been substantiated as fairly close, though purely numerical, approximations over a surprisingly wide range of elements and in diverse phenomena. In their present form they cannot reproduce faithfully the individual characteristics of the various nuclei. Nevertheless, the values of the constants derived may be used with confidence for approximate calculations in many fields of nuclear physics, and for convenient general predictions and comparisons.

The relations utilized and deduced deal solely with the energy of the nucleus as a function of its composition. They might have been expressed equally well if the nuclear composition had been represented by any suitable "components." For this reason the success (or lack of it) of the relations obtained is without bearing upon the truth of the neutron-proton picture of the nucleus. These relations remain, however, as valid approximations in terms of physically real and determinable quantities.

There are two points at which the development may require modification in the future. It is not yet established that the rôles of the neutron and proton as complex and elementary particle should not be exchanged. Secondly, regardless of this, there is some evidence that the beta-ray change is more complicated than is generally assumed. If the existence is admitted of sharp and finite upper limits in the "spectra" of betaparticles, the failure of the conservation of energy in the individual occurrences can no longer be used to explain the variation in energy of the particles. The simultaneous emission of a second particle of some kind must then be assumed. Reformulation of the beta-process would be required in either case if present assumptions prove incorrect.

In conclusion, I desire to express my appreciation of valued suggestions received from Professor Heisenberg, who, in October, 1932, kindly criticized a preliminary and incomplete form of the manuscript of this paper. I have also benefited greatly from discussions with my colleagues.