The effects on the lowering of the vapor pressure of ammonia by thiocyanate and by water have been considered elsewhere[1] and need not be discussed here. It may be noted, however, that equal weights of water and of thiocyanate have approximately equal effect on lowering the vapor pressure of ammonia. For equal numbers of molecules, therefore, thiocyanate has much the greater effect, indicating greater complex formation in solution.

Summary.

1. Vapor pressures at 10°, 20° and 30° have been determined (up to approximately 2 atmospheres) in the system ammonia : water : ammonium thiocyanate.

2. The solubility curves of ammonium thiocyanate in this system have been determined at the same temperatures.

3. The results show that at the temperatures investigated, curves of equal vapor pressure in the ternary system are very nearly straight lines which connect points of equal vapor pressure in the two binary systems.

NEW HAVEN, CONN.

[CONTRIBUTION FROM THE KENT CHEMICAL LABORATORY OF THE UNIVERSITY OF CHICAGO.]

NATURAL SYSTEMS FOR THE CLASSIFICATION OF ISOTOPES, AND THE ATOMIC WEIGHTS OF PURE ATOMIC SPECIES AS RELATED TO NUCLEAR STABILITY.

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1. The Isotopic Number.

Up to the present time no general systems for the classification of isotopes have been proposed,[2] though atomic species have been listed

[1] Foote and Hunter, *loc. cit.*; Perman, *loc. cit.*

[2] However, the listing of isotopes according to their f values, as in Figs. 4 and 5 and Table I, THIS JOURNAL, **42**, 1956–97 (1920), where the f, which is $^1/_2$ the isotopic number, is a term in the Harkins-Wilson atomic weight equation, practically introduced this scheme of classification.

The idea of ordinal or atomic numbers for the elements was first developed by Rydberg in 1896, and was further developed by the remarkable experiments of Moseley in 1913. In the latter year Soddy and Fajans found that some of the radio-active elements exist in isotopic forms. This made it apparent that *to designate an isotope it is necessary to give the values of two variables*, the atomic number and one other variable. The simpler form of the Harkins-Wilson atomic weight equation, $W = P = 2M + 2f$, specified this variable as f, which varies from $-^1/_2$ for hydrogen and 0 for most of the light atoms of even atomic number, up to 27 for uranium. The first plot of this equation was presented by Durrant, in 1917, and the values of f for all of the known elements were published by the writer in 1920. Since f for the complex atoms has all of the half values between 0 and 27, it is evident that by multiplying the f values by 2 they will be changed into whole numbers varying from 1 to 54, and that is what has been done in this paper. In a recent paper (*Phil. Mag.*, **41**, 281–5 (1921)), received after the present paper by the use of their atomic numbers. It is evident that the specification of this one variable is not enough to fix the individuality of an isotope, so it becomes essential to discover at least one other variable which may be used for this purpose. It is true that the atomic weight has thus far been used to specify the individual isotope, for example the three isotopes of magnesium may be listed as Mg 12^{24}, 12^{25}, and 12^{26}, but it is easily seen that this is a method of *indexing* rather than of *classification*, since it can hardly be said that all atomic species of atomic weight 25 constitute a class, especially since at the present time only one atomic species of this sort is known, and even if several such atomic species should exist, they would not be related in stability nor with respect to any other known property aside from mass and gross positive charge.

The first method of classification which will be discussed may be supposed to arise in the following way. Let P indicate the *experimentally determined* atomic weight of a pure atomic species. The experimental methods may consist in the use of the positive-ray method according to the system of Thomson, of Aston, or of Dempster, or of the chemical method, provided in the latter case that the purity of the atomic species is made evident in some other way. Let M denote the Mendelejeff, Rydberg, Moseley element number, usually and improperly designated as the atomic number. This quantity rests on the double experimental basis of the Mendelejeff system combined with the relation to this system of the disintegrations of the radio-active atomic species as expressed by what is commonly known as the rule of Soddy and Fajans, together with the linear X-ray spectrum relation of Moseley. Since both P and M are experimental, their difference, P-M, or N, *is also experimental.* In the language of hypothesis P may be supposed to represent the number of hydrogen nuclei or positive electrons, and N, the number of negative

was submitted, Masson has presented these relations in the form of chemical formulas for the nuclei of atoms. His formulas are not constitutional, but represent only composition, and, while undoubtedly developed independently, are based upon the principles developed earlier by the writer (THIS JOURNAL, **42**, 1956–97(1920)). Thus his fundamental formula $(p_2e)Me_M$ (the symbols are those used by the writer) is merely the expression of the relation $N/P = 1/2$, and $P - M = N$, from which $M = N = P/2$, for elements of zero f value or zero isotopic number. Here e stands for a negative electron, M for the atomic number, and N for the number of negative electrons. The present nomenclature was introduced by the writer in order to use the same letter for the positive electron (p) as he had used in the form of a capital letter (P) in earlier papers to designate the number of positive electrons. For other reasons Rutherford has adopted the same nomenclature (*Phil. Mag.*, **41**, 283 (1921)). It was the intention of the writer to present the relations in the present paper in the graphic form only, but a few equations of the Masson type have been introduced to illustrate the writer's hypothesis that electron groups which contain one negative electron may be stable with respect to disintegration, but unstable with respect to aggregation. These formulas have been evident to the writer for a year, as they are the expression of relations developed at that time. (THIS JOURNAL, **42**, 1960–1, 1964–9, 1970–81 (1920).)

electrons, in the nucleus, while M gives the net positive charge on the nucleus.

A recent paper[1] has shown that the ratio N/P is of fundamental importance in connection with nuclear abundance and stability, since *in no known atomic species is N/P less*[2] *than* $^1/_2$ or 0.5. This ratio increases with increasing values of M, or, in the language of hypothesis, as *the nucleus becomes more positive with reference to its net charge, it becomes more negative with reference to its relative content of electrons.* Thus the stability may be said to be maintained, as its positive charge increases, by a balancing of these two factors.

Though the paper mentioned above considered the relationship between the ratio N/P and M as correlated with nuclear stability, the particular diagram with these two variables independent, as presented here in Fig. 1, was not given. While this plot makes apparent many important nuclear relations, all that will be considered here is the natural system of classification of isotopes which appears. It is evident that each pure atomic species represented falls either upon one of 54 equilateral hyperbolas, or upon the X-axis itself (but below the axis in the case of hydrogen and helium of mass 3) the latter representing a value of 0.5 for N/P. To this, the lower asymptote of all of the hyperbolas, the number zero will be attached, while the hyperbolas, as may be seen, have been numbered from 1 to 54, beginning with the one nearest the asymptotes as 1. The numbers thus obtained, which vary from 0 to 54, will be designated as the *isotopic numbers* of the atomic species. It can be shown that both the stability and the composition of atom nuclei are related to the isotopic number, while there is no such general relation to the atomic weight. Thus the isotopic number classifies the atomic species, while the atomic weight merely serves to index them. As an illustration of the use of these methods of designation the three isotopes of magnesium may be listed as follows: Mg 12_0^{24}, 12_1^{25}, and 12_2^{26}, where the subscript represents the isotopic number, and the superscript, the atomic weight. According to Dempster's experiments ordinary magnesium consists of 75% of isotopic number 0, 12.5% of isotopic number 1, and 12.5% of isotopic number 2. This illustrates in a special case the general relation that about 70 to 80% of all known material is represented by isotopic number 0, though the most abundant isotope of an element has this number in general only when the atomic number is both an even and a small number, not higher than 20. The isotopes of krypton may be designated as 36_6^{78}, the least abundant; 36_8^{80}, 36_{10}^{82}, 36_{11}^{83}; 36_{12}^{84}, listed by Aston as the most abundant isotope; and 36_{14}^{86}, which he considers as the

[1] *Loc. cit.*

[2] The value $^1/_2$ for N/P suggests the formula p_2e, where p represents a positive electron or proton, and e, a negative electron.

second in abundance. Thus the isotopes of krypton may be specified by the isotopic numbers 6, 8, 10, 11, 12 and 14. The maximum abundance is seen to be shifted from isotopic number 0, where it lies for elements 6, 8, 10, 12, 14, 16 and 20; and from isotopic number 4, for elements 22, 24 and 26; to isotopic number 12. It may be noted that the isotopic numbers thus far listed as representing maxima of abundance differ by 4 or a multiple of 4 by a whole number, and that the atomic numbers thus far listed are all even numbers.

The isotopic number may be considered as the number of neutrons (neutron being a term representing one negative electron and one hydrogen nucleus, with a formula *pe*)[1] present in any isotope in excess of the composition represented for the same element by the isotopic number zero. It may be defined in terms of *experimentally* determined quantities by the following equations in which n represents the isotopic number, which has the value of any positive whole number. The hyperbolas are represented by $P(N/P - 0.5) = 0.5\,n$ or $P(2N/P - 1) = n$ so:

$$n = P - 2M \tag{1}$$

$$n = 2N - P \tag{2}$$

$$n = N - M \tag{3}$$

Equation 1 may be written $P = 2M + n$, *so the isotopic number is the number which, when added to twice the atomic number, gives the atomic weight.* Equation 2 indicates that it may also be considered as the *excess of twice the number of nuclear negative electrons over the total number of positive electrons*, and Equation 3 that it represents the excess in the number of nuclear negative electrons over the net positive charge on the nucleus. The value of n is just twice that of the term f in the simpler of the atomic weight equations developed by Harkins and Wilson,[2]

$$W = P = 2(M + f) \tag{4}$$

in which f was supposed to represent the number of β or negative cementing electrons in any atom whose nucleus is built up from α particles and negative cementing electrons alone.

The plot in the lower right-hand corner of Fig. 1 represents the uranium-radium, thorium, and actinium series of radio-active elements. As is well known, the position of the actinium series is still doubtful, since it is not known from just which isotope of uranium, known or unknown, it springs. The plot has been drawn on the basis of the ordinary idea that this series originates in uranium II, marked U_2 in the diagram. If it should prove that it is not correctly located, all that will be necessary to correct it will be to move the whole series up (or down) along the lines of constant atomic number (isotopic lines). It will be seen that the curvature

[1] Here p represents a positive electron or proton and e, a negative electron.

[2] This equation may be written $W = P = 2M + n$.

of the hyperbolas in this portion of the diagram has become so slight as to be almost indistinguishable.

It is evident that during a series of α disintegrations the isotopic number remains constant, while a single β disintegration decreases the isotopic number by 2. Of these 2 units one is due to the *decrease* of N by one, the other to the *increase* of M by one. All of the isotopes of one element (atomic number constant) may be said to lie on isotopic lines, which in the present plot are straight lines which slope upward toward the right, the steepness of the slope decreasing with increasing atomic number. The lines of constant isotopic number may be designated as *neutronal* lines, since each line represents a constant number of neutrons (pe) in excess of the composition $(p_2e)\ M$ represented by isotopic number zero.

The addition of a negative electron to any nucleus would increase its isotopic number by 2, but decrease its atomic number by 1, while the addition of a positive electron would decrease its isotopic number by 1, and increase the atomic number by 1. Thus the addition of a positive electron to the nucleus of Mg 12_2^{26} would give 13_1^{27}, which is aluminum.

Since an isotopic number equal to zero, which represents about $^4/_5$ of all known atoms, corresponds to a ratio of N/P of 0.5, or of P/N equal to 2, any atom nucleus of isotopic number zero may be represented by the formula $(p_2e)_M$, or $(a/2)_M$, while any nucleus may be designated by the formula $(p_2e)_M$, $(pe)_n$, or $(a/2)_M\ (pe)_n$, where a represents an α particle, and the half values of a may be taken to indicate the rarity of atoms of odd atomic and zero isotopic number. It does not seem unlikely that the group consisting of 2 protons and one negative electron (p_2e) may be the most fundamental, but not the most abundant group concerned in atom building. If it is, it is probable that while it is stable with reference to disintegration, it is not stable with respect to aggregation, and the tendency of negative electrons to occur in pairs is such that the p_2e group forms aggregates of the type of $(p_2e)_2$ or the α particle. Li_0^6, B_0^{11}, and nitrogen (N_0^{14}) have *odd* values for M *which accounts for their rarity.*

2. Second System of Classification of Isotopes.

Earlier papers of this series have shown that the elements of even atomic number are about 70 times as abundant as those of odd number, in the meteorites, and about 8 times as abundant in the surface of the earth. It has also been indicated that atoms in which the number of nuclear negative electrons is odd, are relatively rare, that those in which the number of hydrogen nuclei is odd are also not abundant, while the highest abundance is associated with the presence of an even number of electrons.

These considerations suggest that the nuclei of atoms may be divided into 4 classes, depending upon whether N and P are represented by even or by odd numbers, and since both P and N are experimentally determined, the classification is an entirely experimental one.

A consideration of the relations exhibited by Fig. 1 will show that while *all of the abundant species of atoms of isotopic number zero belong to Class I*, relatively rare atomic species of the same isotopic number are present,

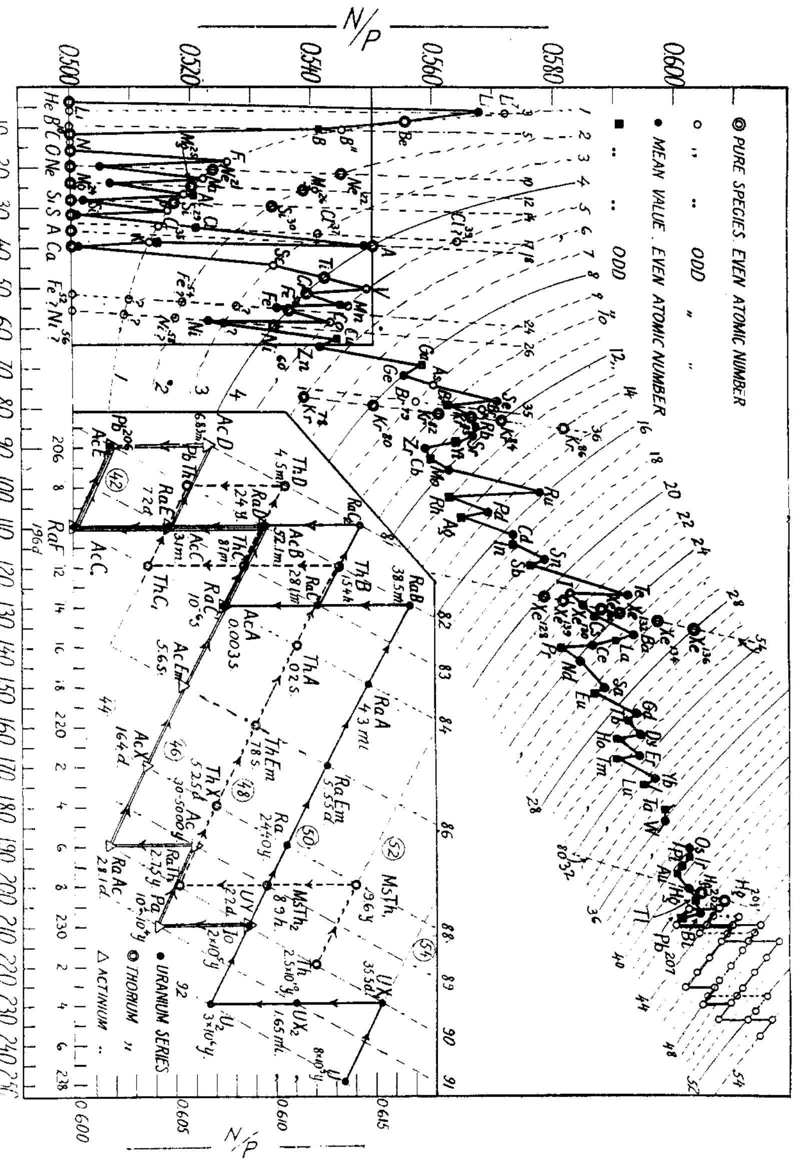

Fig. 1.—The ratio negative to positive electrons in atom nuclei, and the isotopic number.

(Note.—A more recent determination of the atomic weight of antimony by Willard raises its mean position to isotopic number 19.6, thus reducing the greatest known jump in isotopic number between Sb. and I.)

but these belong to Class IV. They are N 7_0^{14}, B 5_0^{10}, and probably Li 3_0^6, though the last of the three has not been found directly.[1]

TABLE I.—CLASSIFICATION OF ISOTOPES ACCORDING TO EVEN OR ODD NUMBER OF ELECTRONS.

		Abundance in atomic percentage in.	
		Earth's crust.	Meteorites.
Class I	N = even. P = even..........	86.9	93.2
Class II.	N = even. P = odd..........	11.2	2.0
Class III.	N = odd. P = odd..........	2.3	3.2
Class IV.	N = odd. P = even..........	0.0	0.0

Even nitrogen, by far the most abundant of these atomic species, is contained in only a small percentage in the lithosphere, atmosphere, and hydrosphere, taken as a whole (only 0.03% by weight and about 0.06% by atomic percentage). It is to be expected that a number of rare species with isotopic number 0 and belonging to Class I, such as A_0^{36}, will be found outside the limits of greatest stability. The important point is that all of the abundant species thus far found for this particular isotopic number belong to Class I.

The abundance of atoms of isotopic number 1 is well distributed between Classes II and III, the atomic percentage for the former class being about 10.5 in the earth's crust and 2 in the meteorites, while for the latter class the corresponding figures are about 2.3 and 3.

All of the known species of isotopic number 2, Ne_2^{22}, Mg_2^{26}, and probably a Si_2^{30}, belong to Class I. Of these Mg_2^{25} is the only moderately abundant one—about 1.5% in the meteorites and 0.21% in the earth's crust.

The only atomic species thus far discovered for which the isotopic number is 3 is Cl 17_3^{37} and presumably also Sc 21_3^{45}, though that scandium is a single species has not been proved. The atomic percentage of the former (Cl 17_3) is only about 0.007%, while that for scandium is so small that it has not been determined. Both of these species belong to Class II.

The known species of isotopic number 4 belong to Class I. Of these iron 26_4^{56} is undoubtedly quite abundant, 12% in the earth's crust and 2% in the meteorites. Ni_4^{60} is probably moderately abundant in the

[1] That lithium consists of the more abundant isotope of atomic weight 7, and a less abundant of atomic weight 6, and boron of the more abundant 11 and less abundant 10, was predicted in a paper sent by the writer to THIS JOURNAL in April 1920 (published Oct. 1920). While the isotopes of boron as predicted were discovered before the publication of the paper, the isotopes of lithium were not found until nearly a year after the prediction was made. *The experiments confirmed the theory exactly since the more abundant isotope proved to have the atomic weight 7, the less abundant, 6.* While it may seem simple to make such predictions, it happens that three other writers made the predictions, and in no case were they completely verified; thus one writer predicted atomic weights for lithium of 6 and 8, another predicted 5 and 7, and a third 6, 7 and 8. For the description of the experiments see a note in *Nature* by Aston (**106**, 827 (1921).)

meteorites, and titanium occurs to the extent of about 0.28% and chromium to about 0.02% in the earth's crust.

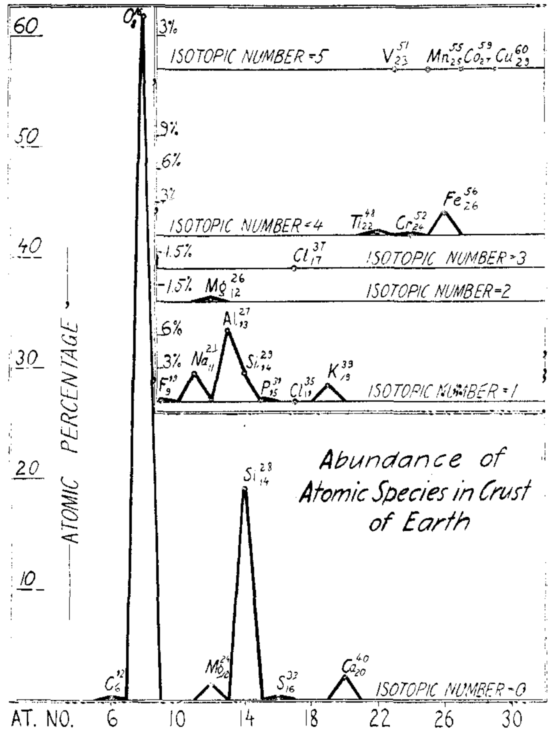

Fig. 2.—The abundance of the pure atomic species (isotopes) as a function of the isotopic and the atomic numbers. It illustrates the periodicity in abundance, and also the great abundance of species of isotopic number zero; and *shows that a secondary maximum, which is very much more prominent in the material of the meteorites, occurs in isotopic number 4*. The figure should be considered as a 3-dimentional plot, with the atomic number on the X-axis, the atomic percentage on the Y-axis, and the isotopic number on the Z-axis. The unequal spacing of the isotopic numbers in the plot was introduced in order to save space. the large space reserved for isotopic number four was necessary in the similar plot concerning the meteorites. The lower isotopes of iron and nickel are not represented, since their atomic weights and therefore their isotopic numbers are uncertain. *The abundance of the atomic species may be seen to be periodic both in relation to the atomic and to the isotopic numbers.*

All of the known atomic species of isotopic number 5 belong to Class II. They are considerably less abundant than those of number 4, but much more so than those of number 3; and about as abundant in comparison with those of number 4, as the species of isotopic number 1 in comparison with those of number 0. Atomic species of isotopic number 6 are undoubtedly considerably more rare than those of isotopic number 5.

Thus the abundance of the species of isotopic number 0 is extremely high, and decreases very rapidly to isotopic number 1, still more to isotopic number 2, with a very low minimum of abundance for isotopic number 3. There is then a moderately high maximum for number 4, after which the abundance again decreases to isotopic number 5, and still again to number 6, so that the maxima occur for isotopic number 0 and the multiples of 4. While in the region of low isotopic numbers these maxima are very high when compared with the abundance of the adjacent isotopic numbers, it is to be expected that they will become much less prominent as this number increases, since the complexity of the nucleus also increases.

The small rectangular plot in the lower left hand corner of Fig. 1 is of interest, since, even although within it are contained the mean value representations for only 27 elements, it contains every one of the 14 most abundant elements on earth, and every one of the 18 most abundant atomic species.

It will be seen that the highest abundance for the species of isotopic number 0 lies between the position for oxygen and that for calcium, that for number 1 it lies between fluorine and potassium, with its maximum in the middle with Na, Mg_1, Al, and Si_1. *The abundance decreases in either direction along the hyperbola; as N/P increases, presumably because such an increase makes the nucleus relatively too negative; and as M (or P) increases, since such an increase makes the nucleus too positive with respect to its net charge.* The position of maximum abundance, and therefore of maximum stability, on each hyperbola, is indicated in a general way by the positions of the points which represent mean values, but cannot be fixed precisely until all of the isotopes, together with their abundance, are known.

It is of interest to note, as is indicated in the upper right-hand corner of Fig. 1, that Hg 80_{44}^{204} is a direct continuation of the thorium disintegration series, and that Hg_{42}^{202} is in the same way a continuation of the radium series. Both of these isotopes have been found recently by Aston. It is not unlikely that the higher isotopes of platinum and osmium will continue one or both of these series still further.

Attention should be called to the fact that the pattern made by the heavy lines used to connect the points representing mean values, as shown in the main portion of the diagram, is not related either to the disintegrative or aggregative processes, but is only intended to point out the periodic nature of such a plot, which is very marked. It is easily seen that the true pattern must be similar to that given for the radioactive elements in the lower right hand corner, though the relations in the main plot will be very much more numerous; for it is apparent that new series, other than the thorium, radium, and actinium series, make their appearance just below the radio-active elements, since atomic species of odd atomic weight and odd atomic number, and of odd atomic weight and even atomic number, which have not been found up to the present time in the radio-active series, exist in all of the other regions.

3. Representation of the Isotopic Number by a Straight Line Plot.

Thus far five variables, P, N, M, n, and N/P, have been used in describing the composition of atom nuclei. It is apparent that any two of these may be taken as the independent variables, when the other three

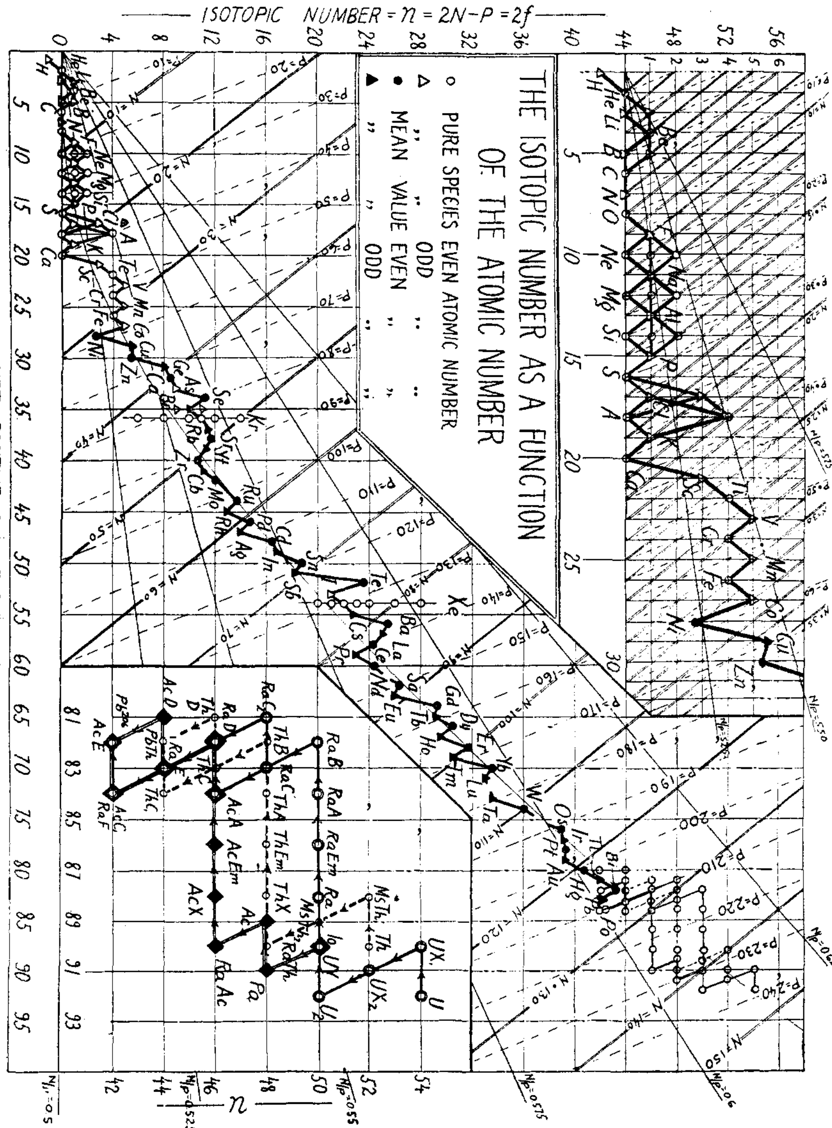

Fig. 3.

(NOTE.—Sb should be higher and at isotopic number 19.6.)

will be dependent. When P and N/P are made independent as in Fig. 1, constant values of n, the isotopic number, are represented by hyperbolas. By choosing n and M as the independent variables it is, obviously, possible to represent constant values of n by straight lines. The remarkable feature of the plot thus obtained, Fig. 3, is that constant values of all of the five variables are represented by straight lines,[1] and that for each of four out of the five, n, M, N, and P, different constant values are represented by a series of parallel straight lines; while various constant values of the ratio N/P are indicated by straight lines radiating from the origin $n = 0$, $M = 0$. It will be seen that the N lines, presumably representing a constant number of negative nuclear electrons, have a slope equal to —1, while the P lines, indicating a constant atomic weight and presumably a constant number of hydrogen nuclei or positive electrons, have a slope of —2. In many other respects this plot is similar to that given by Fig. 1, so that the description need not be repeated, but it will be seen that the line of maximum stability is here changed to approximately an hyperbola, tangent to the M-axis at the origin. In the upper left hand corner of the figure an enlarged plot for the light atoms is given. Although Fe 26_4^{56} is represented as a pure species for iron, it is not to be supposed that this is the only isotope, since the element weight, 55.84, indicates the presence of lower isotopes. Whether beryllium is a pure atomic species has not been determined, and small percentages of isotopes may be found in the case of the other elements represented as pure atomic species.

The figure indicates that Br 35_9^{79}, and As 33_9^{75}, Br 35_{11}^{81} and[2] Kr 36_{11}^{83}; and I 53_{21}^{127} and X 54_{21}^{129}, are pairs of isotopes with the same isotopic number for both members of the pair. However, only in the pair 35_9^{79} and 33_9^{75}, could one of the pair be an α disintegration or aggregation product of the other, since only in this pair does the atomic number differ by 2, and the atomic weight by 4. It will be seen that in such a pair, if the atomic

[1] The equations for these 5 sets of straight lines are

$P = \text{const.}$	$n = 2(q - M)$
$N = \text{const.}$	$n = (q - M)$
$n = \text{const.}$	$n = q$
$M = \text{const.}$	$M = q$
$N/P = \text{const.}$	$n = k\,M$

In these equations q is any positive whole number, and k is a constant which may have any positive value, but is for real cases always a proper fraction with values between zero and some unknown limit which is probably not greater than 0.7 unless in some very exceptional cases. For any known atomic species the maximum value is 0.61, in the case of radium B.

[2] There should also be a Rb 37_{11}^{85}.

number differs by 1, the atomic weight differs by 2, or the difference is represented by the formula p_2e.

It is obvious that one α and two β disintegrations result in a lowering of the isotopic number by 4 while the atomic number is left unchanged. That the maximum of abundance among the light atoms shifts directly from isotopic number 0 to isotopic number 4 is thus strongly suggestive of the hypothesis that the α particle is an extremely important unit in nuclear building. Rutherford has recently obtained evidence of the existence of high-speed nuclei of mass 3 and net positive charge 2, represented by $(p_3e)^+$, which are nuclei of an isotope of helium. While for this particle the ratio N/P is equal to $^1/_3$, which is less than the minimum $^1/_2$ for any known atomic species, no evidence has been presented to show that it has a relatively stable nucleus except with respect to *disintegration*, so it is not known that it corresponds to a real species of atoms. It is not unlikely that when the speed of such a particle is reduced it would either pick up a negative electron, and so be converted into an isotope of hydrogen, or it may pick up a neutron, giving an α particle.[1] Sufficient evidence has been presented in the earlier papers of this series to show that the most important unit in the composition of the light atoms, as well in the heavy atoms, is the α particle. That there should be such a structural unit of mass 3 of secondary importance, is not at all surprising, in fact that such a unit exists was an important part of the theory in its original form. The composition of the complex nuclei indicates that in this *group* it has the composition $(p_3e_2)^+$ which is the composition for an isotope of hydrogen. This does not indicate that such a group could exist for any considerable time or even at all as an independent nucleus, especially since its ratio for N/P is 0.666, which is extremely high for a light atom. The remarkable feature of Rutherford's work is that he seems to get the particles of mass 3 from carbon and oxygen atoms; which would suggest, provided the mass and charge of this particle have been correctly determined, either that some of the α particles in these complex nuclei are broken apart by the bombardment, or that some of these atoms, probably only a relatively small number, have nuclei which are built up partly from particles of mass 3. This question will be discussed in another paper, since it is desired that the present treatment shall be restricted to relations connected somewhat directly with the methods of classification of isotopes under consideration.

Fig. 4 gives an enlarged plot of a small part of Fig. 3 in order to bring out the relations more clearly. Each corner of a square made by the

[1] Thus it may be assumed that the p_3e particle, if it exists, like the p_2e particle, is very difficult to disintegrate, but readily picks up other electrons or electron groups, so it is unstable with respect to aggregation.

vertical and horizontal lines, represents a possible atomic species. It may be seen that if A 18_4^{40} were to undergo a β disintegration it would form K 19_2^{40} which would contain an odd number (21) of negative electrons. It is possible that it is this isotope which causes the radio-activity of potassium, but this may be due to a still higher isotope. The next corner met with in going down the dotted line which represents P equal to 40, is that for Ca 20_0^{40}. It will be seen that on this plot Si 14_2^{30}, Mg 12_2^{26}, Ne 10_2^{22}, and also Sc_3, K_3, and Cl_3 show exactly the relations found in a radio-active α disintegration series among the heavy elements. The

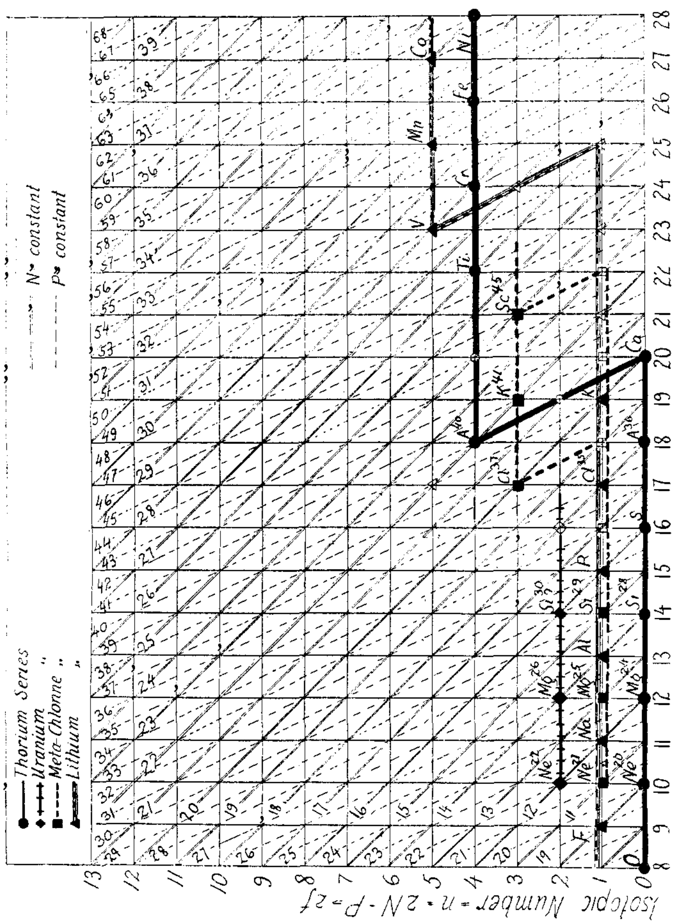

Fig. 4.—The isotopic number plotted against the atomic number. The small squares, circles, and triangles which are not inked in need not represent actual isotopes. They are only added to show the general form of the series relationships.

same is true of K 19_1^{39}, Cl 17_1^{35}, P 15_1^{31}, Al 13_1^{27}, Na 11_1^{23}, and F_1^{19}, and also of Si 14_1^{29}, Mg 12_1^{25}, and Ne 10_1^{21}, and of the long series Ca 20_0^{40}, A 18_0^{36}, S 16_0^{32}, Si 14_0^{28}, Mg 12_0^{24}, Ne 10_0^{20}, O 8_0^{16}, and C 6_0^{12}. It will be noted that on the line representing isotopic number 0 only the corners of even number are occupied, and that the same is true thus far for isotopic number 2, while on the line for isotopic number 1 every corner over the range from atomic number 10 to number 15, is taken. While some of the unoccupied corners may be filled later, the atomic weights indicate that they cannot well represent anything but very small percentages in abundance.

4. Radioactive Disintegration Series.

As is well recognized, the present nomenclature for the radio-active atomic species is extremely irrational and confusing, since it does not sufficiently identify the atomic species. For example in U_{II} the subscript indicates a second isotope of uranium, but whether a higher or a lower isotope is not specified. UX_2, however indicates an atomic species of the element Bv, the third disintegration product in the uranium series, while Io as a symbol shows neither that it is an isotope of thorium, nor what isotope it is, nor even that it is a member of the uranium series. According to the system of classification proposed in this paper, ionium could be designated as $90_{50}^{230}92$, where 90 is the atomic number, 50, the isotopic number, 230, the atomic weight, and 92, the atomic number of its ancestor. Since the atomic weight is twice the atomic number plus the isotopic number, the weight is superfluous, so the designation may be changed to 90–50–92, or since it is the fifth disintegration product, it could be represented by 90–50–92–5. However, since the use of a series of numbers, while more rational, is usually not favored, ionium could be represented as uranio-thorium 50, or UTh_{50}, or if the number in the disintegration series is desired, U_5Th_{50}. Radium would be U_6Ra_{50}; radium emanation, U_7Ra_{50}; radium A, U_8Po_{50}; RaB, U_9Pb_{50}; RaC, $U_{10}Bi_{48}$; RaC′, $U_{11}Po_{46}$; RaD, $U_{12}Pb_{46}$; RaE, $U_{13}Bi_{44}$; RaF, $U_{14}Po_{42}$; and RaG, $U_{15}Pb_{42}$. While these symbols seem more complicated than those in use, this is only because they express so much more. They become quite simple if the number in the series is omitted. Thus UPo_{42} represents uranio-polonium of isotopic number 42. If it is remembered that the atomic number of polonium is 84, then twice 84 plus 42 gives 210, the atomic weight. The isotopic number 42 shows that this atomic species, if it disintegrates, would (unless it were at the point of the branching of a chain, which does not often occur) give off an α particle. That it does this is shown by the fact that the next member of the series, UPb_{42} has the same isotopic number, which is an indication of an α change. β changes in the series are also easily recognized, since, as has been mentioned before, each β change lowers the isotopic number by two, as between UPb_{50}, and UBi_{48}.

5. Number of Isotopes and Atomic Weights.

The first paper of this series gave equations for the atomic weights of the elements, and specified the theoretical weight for the first 27 elements. It may now be seen, since the isotopes for a number of the elements in this region have already been found, that *the atomic weight given by the general equation, is that of the most abundant isotope.* It was found that the atomic weight of the most abundant isotope of an element of even atomic number is itself an even number, while that of the most abundant isotope of an element of odd atomic number, is an odd number. It was also noted that in general the atomic weights of isotopes would in general be most likely to differ by 2, by 4, or less often by some higher multiple of 2. Account should also be taken of the idea[1] that all whole numbers are possible differences, except that the atomic species whose nuclei are unstable structures, do not exist for any considerable period, and are therefore rare. The difference to be expected as the most prominent after the lower multiples of 2 is that represented by one neutron, or 1.

While the multiples of 2 are thus prominent as differences in the atomic weights of isotopes, and therefore in their isotopic numbers, there are some positions in the series in which the general theory indicates that differences of 1 are much more probable. Thus the atomic weight equation gives 7 and 11 as the atomic weights of the most abundant isotopes of lithium and boron respectively. These specific isotopes are designated by values of N/P which are very high for nuclei of low net charge; this indicates that lower, rather than higher isotopes are predicted by the theory. If $^1/_2$ is the minimum value for N/P, then the only possible lower isotopes are Li_0^6 and B_0^{10}. The existence of these isotopes was predicted by the writer in the last paper of this series, and the latter of the two was discovered very shortly afterward by Aston.[2] The purpose of the present discussion is not so much to point out the existence of these isotopes, as to apply the principles just outlined in making a partial prediction as to the relations which will be found to exist among the elements in the region of abundant isotopes, between atomic numbers 28 and 81.

The theory advanced in the earlier papers indicated not only that *atoms* of even atomic number should be much more abundant than those of odd number, but also, as was pointed out specifically by N. F. Hall,[3] that the number of isotopes in elements of even number should be in general considerably greater than in elements of odd number, especially in the region between atomic number 28 and the end of the system (U_{54}^{92}).

[1] Harkins, THIS JOURNAL, **42**, 1960–1 (1920).

[2] Since this paper was submitted for publication the former (Li_0^6) has also been discovered independently by Aston and Thomson, and by Dempster.

[3] N. F. Hall, THIS JOURNAL, **39**, 1616–9 (1917).

All of these principles taken together indicate that between atomic numbers 28 and 81 the element weight, or what is usually designated as the atomic weight, merely a mean value, gives considerable aid in the prediction of the atomic weights of the isotopes of elements of *even atomic number*, since it is apparent that in general most of the *even* numbers adjacent to the mean value will represent atomic weights of actual isotopes, though some of the odd values may also do the same, *but not so often*. Thus the determined mean atomic weight of krypton is known, probably not at all exactly, as 82.92. This would indicate very strongly the existence of isotopes of atomic weights 82 and 84, and the considerable probability of the existence of 80 and 86. While it would not definitely predict any odd number, it is evident that the most probable odd number is that of the mean value, 83. All of these were found by Aston as the atomic weights of the isotopes of krypton, with the addition of 78, which however, seems to be present in only a very small percentage amount. The position of tellurium in the plot indicates that isotope number 24, atomic weight 128, a member of the thorium series, is probably the most abundant, and isotopes 22 and 26, members of the uranium series, are very likely to be present as well as some others which may or may not exist in quantities large enough to permit their detection by the positive-ray method (probably 20 and 28 are present).

Of the isotopes of krypton 8–80, and 12–84 belong to the thorium, and 6–78, 10–82, and 14–86, to the uranium series. Here the first number represents the isotopic number, the second, the atomic weight. This classification into the thorium and uranium series does not involve, though it does suggest, the assumption that these isotopes are actually disintegration products of these two atomic species, but only indicates that they have just the composition which such products would have. Kr_{11}^{83} is the first atomic species of its type to be discovered, for, though it belongs to Class III, of which Mg_1^{25} is a member, it belongs to the lithium series, while Mg_1 is a member of the meta-chlorine series. This is apparent, for the addition of one negative electron to the nucleus would change Kr_{11}^{83} to Br_{13}^{83}, which is a member of the lithium series since its nucleus represents the addition of 19 α particles and 3 pairs of negative electrons to the lithium nucleus. The change of isotopic number from the value 1 for lithium to 13 for bromine is in this case due to these 6 cementing electrons, since each electron corresponds to an increase of 2 in the isotopic number.

The isotope of xenon which Aston considers to be the most abundant is X_{21}^{129}, a member of the meta-chlorine series, while X_{23}^{131} on the other hand is entirely analogous to the krypton of odd isotopic number (11) since it is also a member of the lithium series.

6. Isotopes of Elements of Odd Element (Atomic) Number.

While the relations for the isotopes of elements of even atomic number are thus quite complex, since the isotopes consist of members of both the thorium and uranium series of Class I, and of members of the lithium and Cl_3^{37} or meta-chlorine series of Class III, *the relations for the isotopes of odd atomic number are much more simple, not because the number of possibilities is less, but merely because the number of isotopes which are sufficiently abundant and stable to be apparent, is considerably less.* Thus, among the isotopes of odd atomic number we should expect the most abundant to be members of the lithium series, and the second most abundant to be members of the meta-chlorine series, in both cases members of Class II. Among the radio-active elements we find isotopes of odd element number belonging to both the uranium and thorium series, though such atomic species have an extremely low abundance. They belong to Class IV. In such atoms N is odd, P is even, and M is odd. The only atoms of this type which have thus far been discovered are B 5_0^{10} and N 7_0^{14}, though it seems evident that Li $_0^6$ exists.[1] These atomic species are quite rare, considering their general position in the series which corresponds to what is to be expected for atoms of this class and type.

Thus the relations to be expected from the general theory *in the case of elements of odd atomic number* are: (1) isotopes few in number in comparison with the elements of even atomic number; (2) isotopes mostly of odd atomic weight. In addition to this it is probable at least, so long as the atomic number does not become too high (which tends to level out contrasts of all sorts) that (3) members of the lithium series in general are more abundant than those of the meta-chlorine series.

In accordance with these principles it is to be expected, if the mean, or chemical atomic weight is precisely known, and is very close to an odd whole number. that *the element consists of one pure atomic species,* or else mostly of one pure atomic species mixed with very small amounts of others. The most accurately known element weights of odd numbered elements in the region of abundant isotopes (elements 28 to 81), are bromine, 79.92, iodine, 126.92, and silver 107.88. Thus the mean atomic weight of bromine is 80 within the limits of error. If this were an odd number it would indicate the probability that bromine is wholly or almost wholly one pure atomic species. However, since it is an even number it indicates just as strongly that bromine is a mixture of two isotopes, 79 and 81 in atomic weight, and in nearly equal percentages. The atomic weight of iodine, on the other hand is an odd number, 127, which indicates that it consists of only *one* atomic species especially since the number indicates it as a member of the Li series. This is exactly in accord with

[1] Cf. note 1, p. 15.

what has been found by Aston. The atomic weight of silver, being an even number (very nearly) indicates that it consists wholly or mostly of almost equal parts of isotopes of atomic weights 107, of the lithium series, and 109, of the meta-chlorine series.[1]

The element weight of arsenic, also accurately known, as 74.96, or 75.00 within the limits of error indicates it to be a pure or nearly pure species, which is again in accord with Aston's experimental results, since he finds it to be a single species. Since the experimental work might not detect small percentages of an isotope, it cannot be said to be very much more accurate in this respect than the theory.

It is probable that copper, of element weight 63.57, consists of a large percentage of atomic weight 63, isotopic number 5, a member of the lithium series, and a smaller percentage of atomic weight 65, isotopic number 7, of the meta-chlorine series. Predictions of this nature can be expected to be verified only if the experimental basis is sufficiently accurate. On this basis it may be predicted that gallium, atomic weight 70.1, consists mostly of Ga_7^{69}, of the meta-chlorine series, and of G_8^{71} of the lithium series, and that vanadium, manganese, and cobalt are pure or nearly pure species of atomic weights of 51, 55, and 59, respectively.[2] If the element weights of the heavy atoms were known with sufficient precision, the atomic weights and isotopic numbers of the pure isotopes of the elements of odd element number could be predicted, and it is probable that in almost all cases the most abundant isotopes would be found experimentally to correspond with the predictions. It is, of course, possible, according to the general principles, that 3 isotopes of odd atomic weight may be found in some cases, and thus the prediction would not cover all of the individual isotopes. In individual cases, but probably not often, isotopes of even atomic weight may be found, but in most elements of odd number it is probable that they will be so rare as not to be of importance in considering the abundance relations, even if they are abundant enough to be detected by the positive ray methods in their present state.[3]

[1] The atomic weight of antimony 121.773, as recently determined by Willard and McAlpine, This Journal, **43**, 297 (1921)), indicates that it consists of the isotopes 51_{19}^{121} and 51_{21}^{123}, of atomsic weights 121 and 123, with about 60% of the lower isotope.

[2] On the same basis it may be concluded that each of the elements, sodium, aluminum, and phosphorus, is either a single pure species, or else a single species of high percentage, together with a very small percentage of other isotopes.

[3] Aston's latest results (*Nature*, March 17, **1921**), are exactly in accord with the principles stated in the paper. Thus rubidium of atomic number 37, has an atomic weight 85.45, which indicates according to the theory, that it should consist of a mixture of *two* isotopes whose atomic weights are the two nearest *odd* whole numbers above and below 85.45. These are 85 for the more abundant (Rb 37_{11}^{85}) and 87 for the less abundant (Rb 37_{13}^{87}) isotope, which is exactly what was found by Aston. The atomic weight for potassium, 39.10, if sufficiently exact, would indicate that this element should contain

The principles listed above make it possible to make a rough estimate to be made of the number of isotopes which will be discovered for the 86 elements now known, provided no new isotopes in the radio-active region are found, and also that all of the 86 elements are investigated by forms of the positive-ray method which have the same sensitiveness as those used during the last year. The number of atomic species to be expected lies in the range from 280 to 300. A sufficient increase in the sensitiveness of the methods would probably increase this number considerably above 300, since there is much ground for the prediction that many atomic species exist in such small amounts as not to be capable of detection by present methods.

It is evident from Fig. 2 that there are as many *possible* isotopes of one complex element as of another if only the isotopic numbers are taken into account. Since the equation characteristic of isotopes is $P - N = M$ it is apparent that a small number for M does not of itself make P small. In addition to this it seems almost certain from the discussion presented earlier in the paper, that the most stable isotopic species are among those in which M is small. From this alone it might seem that there should be more isotopes for elements of low than for those of high atomic number, which is nearly the opposite of the truth, for, even though the highest peaks of stability lie in the region where M is small, the relative change in composition produced by adding or subtracting only one or two neutrons, is so great as frequently to throw the atomic species into the region of extreme instability. When, on the other hand M is large, especially since $N \overset{=}{>} M$, and $P = N + M$, the number of particles is so large that the addition of one or two neutrons produces a much smaller relative

about 95% of the isotope K 19_1^{39} and 0.05% of K_3^{41}, which accords with Aston's results. If the mean chemical atomic weight, on the other hand, is incorrect, the principles developed by the writer cannot be expected to indicate the correct atomic weights, any more than the positive-ray method should be expected to give the correct results when carried out inaccurately. Thus the mean chemical atomic weight of cesium, 132.81, indicates that it is mostly Cs_{23}^{133}, but that it also contains about 0.09% of a lower isotope 131, which Aston does not find. Since the whole number rule is much more exact than this we are forced to the probable conclusion that the chemical atomic weight is too low, or else, what seems less probable, but is quite possible, that Aston failed to find the lower isotope, even though it is present. As is indicated by the chemical atomic weight, sodium was found to be a single pure species. The most important fundamental predictions used in the prediction of isotopes of either odd or even atomic number are that N/P is never less than $1/2$ (this was the basis for predicting the 6 and 10 isotopes of lithium and boron, respectively), that the number of nuclear negative electrons is almost always even, and that the value of N/P cannot vary far from the "normal" value without causing the atom to be unstable. Fourth, that occasionally an atom of even atomic number may contain an odd number of both positive and negative electrons. If the present chemical atomic weights of the elements of odd atomic number were entirely exact, almost all of the moderately abundant isotopes could be predicted with very few errors.

effect. However, as the net positive charge on the nucleus (and the atomic number) increases, the stability of the most stable isotope decreases, so, even although the variation in the stability produced by the addition or subtraction of neutrons still decreases with the atomic number, there must be a region for extremely high atomic numbers, in which the number of isotopes again decreases as the atomic number rises. If it were not for some such effect as this, the number of isotopes should rise indefinitely, and be higher for elements of atomic number above 92, than for elements below this, which is what would be expected from the theoretical treatment of Aston,[1] who considers the number of actual configurations to rise with the atomic number. This is directly contradictory to the principle developed by the writer, which indicates that the number of isotopes first increases, reaches a general maximum, and again decreases, as the atomic number increases. The position of this maximum is not yet determined, but it seems probable that it lies somewhat below (in atomic number) that of radio-active elements.

A plot in which the Y-axis represents the number of isotopes abundant enough to be detected by positive rays and the X-axis the atomic number would probably begin with 1 atomic species for He rise to 2 for Li and B, fall to 1 for C, N, O and F, rise to about 3 for even atomic numbers between Ne and Si, with less than 3 (only 1 abundant) for odd atomic weights in this range. Beyond argon and up to iron is probably a region of few isotopes, while beginning with Ni the region of abundant isotopes begins. The plot is markedly periodic, being relatively low on each odd atomic number, except in the regions of lithium and boron and adjacent to the chlorine (with chlorine the isotopic number begins to rise). Where the general maximum lies in this region, it is difficult to see, but possible well up toward the radio-active elements. While we know a considerable number of isotopes in the radio-active region, this is true only because our methods of detection are so very much more delicate for such atomic species. The periods of disintegration indicate that above atomic number 82 there are few elements in which more than one of the isotopes could be detected by positive-ray methods. There is every indication that the number of isotopes has here passed its maximum. Thus the plot is not only periodic with reference to odd and even, but there are also more extended maxima and minima, aside from the highest maximum. The most marked minima seem to be in the region of C, N, O, F, and of Ti, V, Cr, and Mn.

The number of isotopes of low atomic number is made very small too by the precipitous decrease in stability below the line representing N/P = 0.5, or below isotopic number zero. Fig. 1 is especially valuable in considerations concerning the number of stable nuclei for elements of low

[1] *Sci. Progress,* **15,** 212–22 (1920).

and of high atomic number. It shows, for example, that the addition of one neutron to an α particle would lift the representation in the plot to isotopic number 1, with a value of N/P equal to 0.6, which is far out of the region of stability. Both Fig. 1 and Fig. 3 become much more useful if they are considered to represent the X–Y plane, and if either the atomic stability, or the abundance of the atomic species is represented on the Z-axis. Plots of this nature have been prepared by the writer, and their general features have been described in connection with the consideration of the isotopic numbers. In this connection it should be kept in mind that the small rectangle in the lower left-hand corner of Fig. 1, plots all of the abundant atomic species, representing as it does about 99.9% of all of the material on the surface of the earth and in the meteorites.

Summary.

1. An earlier paper classified the different atomic species in series according to the composition of the nuclei of their atoms: (1) thorium, (2) uranium, (3) lithium, and (4) meta-chlorine (Cl 17_3^{37}) series. The present paper adds two natural and experimentally determined methods of classification, which are complementary (1) according to the *isotopic number* and (2) according to the class number as defined in a later section of the summary. It is found that five variables, P, the atomic weight, or total number of protons in the nucleus; M, the atomic number, or the net positive charge on the nucleus; N, equal to P–M, the number of negative nuclear electrons: N/P, which may be considered as the relative negativeness to positiveness of the nucleus; and n, the *isotopic number.* It is evident that these five variables would give ten 2-dimensional plots. but the representation is greatly simplified by the fact that by a proper choice of the two independent variables (n and M) constant values of all five may be represented by straight lines (Fig. 2).

2. The isotopic number represents the number of neutrons of the formula pe necessary to represent the excess in the composition of the nucleus over what may be considered as the normal composition represented by the isotopic number 0, which may be given as $(p_2e)_M$. The isotopic number is exactly twice the value of the function f in the Harkins-Wilson equation for atomic weights, or $W = 2(M + f) = 2M + n$. The atomic species were classified according to their f values in an earlier paper. The isotopic number 0 seems to represent the lowest isotope which is stable both with respect to disintegration and to aggregation, and includes about 70 to 80% of all known material. However all isotopes with this isotopic number are not supposed to be stable. The isotopic numbers higher than 0 are represented by 54 hyperbolas when N/P and P are the independent variables (Fig. 1) and by 54 straight

horizontal lines when n is plotted on the Y-axis. It may be seen that the composition of any atom is $(p_2e)_M(pe)_ne_M$.

3. The relative abundance of the atomic species of the different isotopic numbers is

Isotopic number.	Earth.	Meteorites.
0	84.5	79.0
1	13.0	5.3
2	0.2	1.6
3	0.007	0.0
4	2.2	12.4
5	0.046	0.1
6	Less than for isotopic No. 5.	

Thus the abundance is very high for isotopic number 0, decreases to a minimum in isotopic number 3, rises to a secondary maximum in 4, and again decreases to 5 and 6. The difference of 4 isotopic numbers between the maxima corresponds to 4 neutrons or a helio group, which is an α particle plus two electrons, so it indicates that in this range as well as in the radio-active region, the α particle is an important unit in atom building.

4. In an α disintegration the isotopic number remains constant, in β disintegrations it is lowered by 2. Beta disintegrations of atoms of odd atomic number are in general much more violent than those of even isotopic number.

5. Nuclei may be classified into the four following classes which bear an important relation to nuclear stability: I, both N and P even, comprising about 90% of known material; II, N even and P odd, 5%; III, both N and P odd, 2.5%; and IV, N odd and P even, 0.0%, where all percentages are atomic. Therefore P-N or the atomic number is even for most atoms, and the atomic weights of elements of odd atomic number are almost always odd, while those of even atomic number are usually even.

6. The number of isotopes of elements of even atomic number should be, according to the earlier theory of the writer, considerably larger than the number of isotopes of elements of odd atomic number. On the basis of the relations in this and the preceding sections, the existence of a number of undiscovered isotopes is predicted.

7. The general theory indicates that the number of isotopes per atomic number, reaches a maximum somewhere in the region between atomic numbers 28 and 83, and that there are fewer isotopes in the radio-active region, and very many less in the region of few isotopes between atomic numbers 1 and 27.

8. A rational system for the nomenclature of the radio-active atomic species is proposed.

9. The atomic weights of isotopes of odd atomic number are almost always odd numbers, in order to make the number of negative nuclear electrons an even number. However, the values of the N/P ratio for lithium of atomic weight 7 and boron of atomic weight 11 *are so high* that the presence of lower, rather than higher isotopes is indicated by the theory, thus indicating that lithium has an isotope of atomic weight 6, and that boron has an isotope of atomic weight 10. While these isotopes have been discovered, the writer's prediction of their existence was made in an earlier paper before their discovery. The existence of isotopes of these elements with still lower atomic weights than these would be contrary to the general rule that N/P is never less than $^1/_2$ for stable complex atoms. The principles developed in earlier papers by the writer indicate also that isotopes of even atomic number should have atomic weights which are mostly even numbers, though some odd atomic weights may occur. Thus most isotopes differ in atomic weight by 2.

10. It is suggested that groups such as the p_2e group, which contain *one* negative electron (an odd number) may be extremely stable with respect to disintegration, but that they tend to combine with each other and with other similar groups, or even with negative electrons alone, to form more complex groups in which the number of negative electrons is even. Thus two p_2e groups would combine to form an α particle, or one p_2e group would add on a negative electron to form a p_2e_2 group, or double neutron. Thus such groups, including Rutherford's p_3e group of its exists, would be stable with respect to disintegration, but not with respect to aggregation. In exceptional cases a p_2e group might attach itself to a larger atom nucleus—thus forming the isotope of lithium of atomic weight 6 by combining with one α particle, that of nitrogen by combining with three α particles, or that of boron by a union with two α particles.

There are a number of facts which suggest that the disintegration of an atom need not be the exact reversal of its building up.

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