by the tensimetric method; and that early readings in a tensimetric measurement are higher than later ones.

(2) The suggested explanations of Tammann, Nernst, Campbell, Partington and Brereton Baker have been referred to.

(3) A number of criticisms, which it is hoped may be helpful to other workers, have been offered of the experimental work of users of both tensimetric and gas-current saturation methods.

(4) An experimental re-examination of the facts has been described.

(5) It has been shown that the real facts exhibit no anomaly.

(6) Reason has been given for accepting the tensimetric results of Frowein, often regarded as standard, only with caution.

PRINCETON, N. J.

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

THE STABILITY OF ATOMS AS RELATED TO THE POSITIVE AND NEGATIVE ELECTRONS IN THEIR NUCLEI, AND THE HYDROGEN, HELIUM, H_3, H_2 THEORY OF ATOMIC STRUCTURE.

BY WILLIAM D. HARKINS.

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The purpose of this article is to extend the theory presented in the earlier papers[1] of this series, the first of which were published in 1915. Using as a basis the evidence obtained from the electrolysis of solutions, from the positive ray work of Thomson,[2] and from the atomic weights of the elements and of the separate atomic species, insofar as the latter were then known, the writer presented the theory that the nuclei of all atoms are made up of positive and negative electrons, and evidence was presented which indicated that the nuclei of hydrogen atoms are positive electrons. It will be pointed out later in the present paper that in the light nuclei there are in general about half as many negative as positive electrons, and that in no known complex nucleus is the ratio of negative to positive less than one to two.

The theory indicated that the deviation of the atomic weight of helium (3.969 when calculated on the basis of hydrogen as 1) from a whole number, is due to a "packing effect" which amounts to a decrease of mass equal to 0.77%. This theory was based on the idea of Rutherford[3] that the nucleus of the atom is very minute. That there is a decrease of

[1] THIS JOURNAL, 37, 1367–1421 (1915); 38, 186–214 (1916); 39, 856–879 (1917); 41, 970–992 (1919); *Phil. Mag.*, 30, 723–734 (1915); *Science, N. S.*, 46, 419–427, 443–448 (1917); 50, 577–82 (1919); *Proc. Nat. Acad. Sci.*, 1, 276 (1915); 2, 216–224 (1916); *Phys. Rev.*, 15, 73–94 (1920); *Z. anorg. Chem.*, 97, 175 (1916).

[2] Thomsen, "Rays of Positive Electricity," Longmans, Green and Co. (1913).

[3] Rutherford, *Phil. Mag.*, 21, 669 (1911); 26, 702 (1913); 27, 448 (1914).

mass when a positive and a negative electron approach each other is an essential postulate in the electromagnetic theory of Lorentz,[1] but he did not give the magnitude or even the sign of the effect. This was calculated for us by Professor A. C. Lunn, whose results showed that the packing effect found in the formation of helium from hydrogen, could be accounted for in the simplest system, consisting in one positive and one negative electron, by their approach until the distance between them is about 400 times the radius of the positive electron. In the first paper it was stated that Rutherford considers the hydrogen nucleus to be the positive electron, this information having been given to us in an oral report of one of his addresses. Reference was not made to an important specific part of one of Rutherford's papers which appeared while our papers were in process of preparation, though a general reference to the paper as a whole was given. This specific reference was of importance, and would have been given except for the fact that the writers, in looking over about 150 papers and books, failed to read these 2 pages near the end of a somewhat long paper. The content of these pages cannot be given in full, but it is desirable that the following 2 quotations be presented.[2]

"The exceedingly small dimensions found for the hydrogen nucleus add weight to the suggestion that the hydrogen nucleus is the positive electron, and that its mass is entirely electromagnetic in its origin."

"For the dimensions of the positive and negative electrons considered, the packing must be very close in order to produce an appreciable alteration in the mass due to this cause. This may, for example, be the explanation for the fact that the helium atom has not quite 4 times the mass of the hydrogen atom."

The first two papers of this series were written largely for the purpose of giving the evidence which existed in the atomic weights in favor of the above assumptions, and to indicate that where the atomic weights were not in their favor, the apparent discrepancy could be explained by the existence of isotopes. It was assumed in this connection that not only neon, as found by Thomson, but also chlorine, silicon, magnesium and nickel, copper, zinc, mercury, and nearly all other elements of atomic numbers 28 to 80, as well as the radioactive elements, are mixtures of isotopes. The ordinary atomic weights when considered in a systematic manner, give strong evidence in favor of this assumption.

Four Series of Atoms from the Standpoint of Composition are Now Known.[3]

Atoms belonging to at least 4 more or less independent series are now known, and a fifth, but dependent series also exists. These series are,

[1] H. A. Lorentz, "The Theory of Electrons," 1909, pp. 47 and 48.

[2] Rutherford, *Phil. Mag.*, **27**, 494–5 (1914).

[3] These series of atomic species have been classified according to the *composition*

1. Helium, or Helium-Thorium Series [(a) pure α-series (b), α-cementing electron series].

2. Meta-neon-Uranium Series (α,μ-series).

3. Lithium-Cobalt Series (α,ν-series).

4. Meta-chlorine series (α,ν,μ-series).

The dependent series is,

2b. Actinium series (Secondary α,μ-series), presumably one branch of the Uranium series.

While there are only 92 chemical elements in the series of elements now known, that is only 92 different kinds of atoms, if the arrangement of the outer or planetary electrons alone is considered, it is probable that at least three or four hundred elements exist if the viewpoint taken is that of atomic evolution and disintegration. However, since it is customary to class all elements which have the same planetary electron number and arrangement as one element, the different elements of this type will be considered as *different atomic species*, and the term element, which thus becomes entirely arbitrary, will be used to indicate a chemical element. Each of the 5 series of atoms, when considered in this sense, does not consist of a number of elements, but of a number of atomic species.

The 4 series of atoms are listed above in decreasing order of abundance. Thus *by far the greatest part of all known material belongs to the alpha or helium series.* The nuclei of all the more common atoms of this series may be assumed to consist of *α-particles alone*, or of *α-particles plus negative α-cementing electrons.* For convenience these will be referred to as Class 1 and Class 2 of this series. Most of the abundant atoms, as oxygen, α-magnesium, α-silicon, calcium and probably an α-nickel, belong to Class 1, though iron is very abundant and contains 2 cementing electrons in its nucleus. *No abundant species of atoms contains more than 2 such electrons.* The prefix α indicates a species of atoms whose nuclei consist of α-particles. Each α-particle is assumed to consist of 4 positive and 2 negative electrons (Fig. 1).

The atoms of the meta-neon-uranium series seem to have nuclei which consist of α-particles mostly, but which contain in addition one other group. Since the nature of this group has not been determined experimentally, its exact size is uncertain. However, the difference between a member

of their atoms, and from this standpoint they need not be considered as making up such series as are considered in the disintegration of the radioactive atoms, or even as series from the standpoint of atom building. However, since all of the descendents from uranium show the same type of composition, and since all of the known atoms of this type of composition among the radio elements, have also been proved to be descendents from uranium, it does not seem to be improbable that the series listed above are concerned in atomic aggregation and disintegration. This does not mean, of necessity, that in the building of atom nuclei there can be no passing over from one series into the other.

of the α- or helium series and a corresponding member of the meta-neon-uranium series consists in the presence in nuclei of the latter type of a group consisting of 2 positive and 2 negative electrons, of the formula $(\eta_2{}^+\beta_2{}^-)^\circ$, where η represents a positive and β a negative electron and $^\circ$ indicates that the net charge on the group is zero. *If this group exists by itself, it is a particle belonging to an element of zero atomic number.* It will be called the *mu* group, so the series is an α-μ-series. The lightest particle now known, which contains this group, is the nucleus of the meta-neon atom, which may be assumed to consist of 4 α-particles and one μ-group. It is possible that the primary group from the standpoint of structure may have the composition $(\eta_6{}^+\beta_4{}^-)^{++}$, in which case it would be the nucleus of a meta-helium atom, but the assumption of the presence of the μ-group leads to a simpler system. The atoms of this group, as a class, are not so abundant as those of the α-series, but they are probably more abundant than those of the third and fourth series, at least this will be found to be the case if the highest atomic weight isotopes of magnesium and silicon belong to this series, as is probable and if the present atomic weights[1] of these elements are correct within 0.1 unit. These isotopes have not been discovered, but they exist if the present atomic weights of magnesium and silicon are correct to within 0.2 unit in the case of magnesium and to 0.05 unit in the case of silicon.

Lithium-Cobalt Series.—This series as it is now known begins with lithium and ends with cobalt. It is practically certain, however, that cobalt is not the highest member of the series, and that it extends as high as *gold*, and possibly to bismuth. No members have been discovered among the radioactive elements of atomic number 84 or higher. The atoms of this series make up only about 1.3% of the material of the meteorites, while, so far as is known, about 98% or more belongs to the first 2 series, the helium and the uranium series. While the lithium nucleus is the lightest known particle belonging to this series, it is possible that a nucleus of mass 3 (or ν particle) exists, though it is probable that no appreciable amount of this element can be found on earth.

In the first paper of this series it was considered that the atoms of the lithium series contain a particle of mass 3, which was designated as the H_3 particle. If such a particle exists, though it contains 3 positive electrons or nuclei of hydrogen atoms, it does not contain 3 negative electrons, but less than 3. In the later papers the symbol ν has been used to indicate a *group* of 3 positive and 2 negative electrons $(\eta_3{}^+\beta_2{}^-)^+$, which is very evident in the *composition* of atoms of the lithium series. Whether this *group* contains the same number of negative electrons as the corresponding *particle* of weight 3, cannot be deduced from the general theory, though the only possibilities are that the 3 particles, if it exists, shall contain one

[1] Mg = 24.32, and Si = 28.11.

or two negative electrons, though the indications are that it contains two.

This group is the first to be considered here which has an odd atomic weight, and as a *group* it has an odd positive charge. It will be shown later that it is probable that this *group* is responsible for the odd charge of most light atoms of odd atomic number.

Meta-chlorine Series.—Recent results obtained by Mr. C. E. Broeker and the writer in an investigation started early in 1916 seem to indicate that they have actually separated the element chlorine into isotopes, and the positive ray analysis of Aston indicates that the atomic weights of the isotopes are 35 and 37. Now Cl_{37}, or meta-chlorine, is a member of none of the 3 series already listed. While the nature of the groups present in its nucleus may seem somewhat in doubt, it is the first atom of odd nuclear charge whose atomic weight is 2 higher than the theoretical value, which is 35. Thus meta-chlorine differs from chlorine by the same amount as meta-neon differs from neon, and by the same amount as any member of the α,μ-series differs from the α-series. The simplest assumption is that the nuclei of Cl_{35} and Cl_{37} differ by one μ group, so if the chlorine nucleus is $(\alpha_8{}^{++}\nu^{+})^{17+}$, the meta-chlorine nucleus would be $(\alpha_8{}^{++}\nu^{+}\mu)^{17+}$. While the form of the latter might be $(\alpha_9{}^{++}\eta^{+}\beta_2{}^{-})^{17+}$ the former formula is in better accord with the general system of structure which is found. In either case, however, meta-chlorine belongs to a fourth series.

A study of the atomic weights of the 4 series indicates that in the *helium-thorium* or *α-series the atomic weights are divisible by 4, in the meta-neon-uranium or α-μ series they are divisible by 2 but not by 4*, or whenever divided by 4 a remainder equal to 2 is found. In the *lithium-cobalt series a remainder equal to 3* is left after a division by 4, *while in the meta-chlorine series*[1] *the remainder is equal to 1*.

While the above list may seem to exhaust the list of possible remainders, there are 2 types of atoms which have not been included in the 4 classes or series given above.

Groups which are Responsible for the Existence of Isotopes. (Weights 4, 3, 2, and 1.)

Since the nuclei of isotopic atoms have the same net nuclear charge, their differences must be expressed by groups which have a zero net charge. The principal groups which are now evident are the μ or $(\eta_2{}^{+}\beta_2{}^{-})^{\circ}$ group, and the $(\eta_4{}^{+}\beta_4{}^{-})^{\circ}$ or $(\alpha^{++}\beta_2{}^{-})^{\circ}$ group. The latter is an α-particle together with 2 negative *cementing* electrons, which serve the purpose of attaching an *extra* α-particle to a nucleus, that is an α-particle which is apparent in the mass, but is not apparent in the charge, of the nucleus.

[1] While meta-chlorine was the only member of this series known when this paper was first written, Aston has just reported a xenon of atomic weight 133, which may be either of the type of beryllium, or of the type of meta-chlorine.

It is these cementing electrons which are given off in the β disintegrations of the radioactive atoms. The latter group has the same formula as a helium atom, but undoubtedly differs in that it has a much more compact structure. It will be called a helio group (or particle). All known differences between isotopes of *radioactive* atoms of high atomic weight seem to be caused by one μ group, one or 2 helio groups, or one μ and one helio group. Since members of the lithium series have not been discovered among such radioactive atoms, it is evident that *in this range* no isotopes which differ by a ν group plus a negative electron $(\nu^{+}\beta^{-})^{\circ}$ or $(\eta_3{}^{+}\beta_3{}^{-})$, are known. Isotopes which differ by a ν group plus one negative electron are to be expected in considerable numbers in the range between atomic numbers 29 to 79, and possibly higher. *Ordinary lead* may be either a mixture of uranium and thorium lead, or it may be an individual isotope, and in the latter case it *may* be a lithium derivative. However, it will be shown later that certain rules of structure which seem to hold generally, would indicate that a lithium derivative of even atomic number should have a relatively short life, which makes it seem probable that ordinary lead does not belong to the lithium series. It is not improbable that some isotopic atoms are formed by the addition of the group $(\eta^{+}\beta^{-})^{\circ}$.

The Stability and Building of Atom Nuclei.

The large loss of mass (0.77%) in the formation of an α-particle from 4 positive and 2 negative electrons, indicates a probable structure for the α-particle, since to give this effect the positive and negative electrons should lie close together, while the positive electrons should be apart from each other, as should the negative electrons; since the closer the approach of the positive to the negative electrons, the greater the loss of mass, while the closer the approach of positive to positive or negative to negative, the greater the increase of mass. Fig. 1 presents a model for the α-particle in which the negative electrons, while represented as rings or discs, are not assumed to have any specific form, since the purpose of the figure is to give only a suggested relative position in space of the positive and negative electrons. If both the negative and the positive electrons are spherical, then the electro-

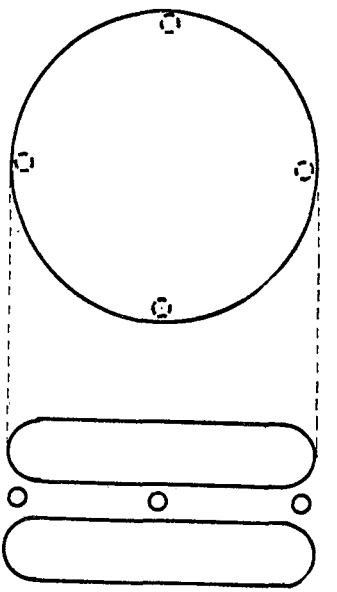

Fig. 1.—Model to illustrate suggested arrangement in the α-particle of positive electrons (small spheres), and negative electrons (large spheres or discs). The model is not intended to suggest any special form for the electrons themselves.

magnetic theory indicates, as has been pointed out by Rutherford, that the radius of the positive is only $^1/_{1835}$ that of the negative. Such questions as why the negative and positive electrons do not neutralize each other, why the apparent number of negative electrons is equal to the number of positive electrons, and why the charges are numerically equal, though of opposite sign, need not be considered here. The difference in size may have some bearing on the first of these questions.

The model suggested should represent an extremely stable system, provided the electrons of opposite sign do not coalesce. It has been suggested by Richardson and by G. N. Lewis that electrons have around them a field of force which alternates in sign as they are approached. On the basis of their theory each electron should consist of a series of concentric shells, decreasing in density with the distance from the center. Such a theory does not decrease the difficulties involved in explaining the stability relations, since it introduces a form of electron so complex that these difficulties are simply transferred to considerations concerning the electron structure itself. According to their theory there might well be a union and a consequent loss of mass with either positive electrons alone, as was assumed by Nicholson, or with negative electrons by themselves. It is of importance in this connection that in considering the composition of any group in which such a packing effect is found to exist we are forced to the conclusion that it consists of both positive and negative electrons, and usually contains more of the former, as in the case of the α or the ν group.

Having adopted a suggestive model for the α-particle based on the idea that electrons of unlike sign should lie close together, and those of like sign relatively far apart, in order to give the packing effect, let us consider the way in which α-particles unite with each other. The early papers of this series advanced the theory that from 3 to 8 and 10 α-particles unite with each other to form complex nuclei, without the inclusion of any other positive or negative electrons. It is probable that 9, and more than 10 α-particles, *probably as many as 14*, also unite, but in none of these cases, unless in nickel with 14, are there enough combinations of this kind to make these nuclei relatively abundant. All of these nuclei belong to the pure α-division of the helium series. Every α-particle carries a net positive charge of 2, so at all distances of ordinary magnitude α-particles would repel each other if at rest, according to the ordinary laws of electrostatics. However, since α-particles unite with each other to form complexes so stable as the nuclei of atoms of oxygen, magnesium, calcium, etc., it is evident that at the very minute distances between the α-particles in these nuclei they must attract each other in spite of their net positive charge. It is evident that this attraction might be considered on the basis of an alternating field around electrons according to the hypothesis

of Richardson and Lewis, but it seems much more simple to treat it as due to the action of electrical couples, which might easily become more powerful at such short distances than the repulsion due to the net positive charge.

The fact that isotopic forms of ordinary elements are now being discovered, and the probability mentioned earlier in this paper that hundreds of such isotopes exist, make it unsafe to state that any special form of union is non-existent. For example, it is not certain that there is no lithium atom of atomic weight 5 though its existence is improbable. The nucleus of such an atom could consist of an α-particle plus one positive electron. An isotope of hydrogen might have a nucleus consisting on one α-particle plus a negative electron. However, it may be stated that if such nuclei exist at all, their quantity is so small that they are of little relative importance. So, when it is stated that neither a positive nor a negative electron make a stable union with an α-particle, the statement is made with reference to the above considerations. The model suggested for the α-particle, which may not be the only model which will meet the conditions, may be said to repel the positive electron on account of its net positive charge, and to repel the negative electron at very small distances, because the outer part of the model is largely negative. At considerable relative distances negative electrons would be attracted.

Since as many as 10, and probably even as many as 14, α-particles unite alone to form a stable intra-nuclear compound, the question may be raised as to why larger numbers of α-particles do not unite in the same way. The nuclei of carbon, oxygen, α-neon, α-magnesium, α-silicon, and sulfur, have, respectively, atomic weights of 12, 16, 20, 24, 28 and 32, and net positive charges 6, 8, 10, 12, 14 and 16. Their atomic weights and net charges are, therefore, 3, 4, 5, 6, 7 and 8 times the weight and charge of the α-particle, so these last numbers may be taken to indicate the number of α-particles in the respective nuclei. The ordinary argon nucleus, however, has a weight of 40, but its charge is only 18, so from its weight it should contain 10, while its charge indicates the presence of only 9, α-particles. The obvious explanation is that this nucleus contains one α-particle which is apparent in the weight, but is not apparent in the charge. That is, the argon nucleus contains enough extra or alpha *cementing* electrons to neutralize the positive charge of one α-particle. The number of α-cementing electrons rises as the atomic weight (number of positive electrons), or the number of α-particles increases, until in the thorium nucleus there are 26, and in lead from thorium, 22 such electrons. Argon is the element of lowest atomic number in whose normal atom these cementing electrons appear. *Thus, in the lightest atoms there are no α-cementing electrons, in the heavy atoms there are no nuclei without α-cementing elec-*

trons. The question which now arises relates to the necessity for the inclusion of these cementing electrons, together with the *extra* α-particles which always accompany them, in order to give stability to a heavy nucleus, while the stable nuclei which contain from 3 to 8, and even 10, α-particles, contain no such cementing electrons.

Suppose that several α-particles are approaching each other. Their mutual self-repulsion due to their net positive charges, tends to keep them apart unless they are driven together so strongly that they approach sufficiently close to enable the attractive effects of the mutual couples to become of significance. As the total net positive charge on the nucleus, once it is formed, increases, the repulsion due to this cause may be assumed to increase to such an extent that α-particles are no longer able to pass through the region of repulsion, and to attach themselves, though electrically neutral particles, such as the helio group $(\eta_4{}^+\beta_4{}^-)^\circ$ or $(\alpha^{++}\beta_2{}^-)^\circ$ group the mu group $(\eta_2{}^+\beta_2{}^-)^\circ$, the $(\eta_3{}^+\beta_3{}^-)^\circ$ or $(\nu^+\beta^-)^\circ$ group, and the $(\eta^+\beta^-)^\circ$ group could easily pass into and through this region.

The ratio of the number of negative to the number of positive electrons in the α-particle (helium nucleus) is one to two, or 0.5, and this may be said to be also the *normal* ratio in light atoms whose nuclei are made up from α-particles alone. This is the exact ratio in the nuclei of carbon, oxygen, α-neon, α-magnesium, α-silicon, sulfur and calcium; and also in α-argon (at. wt. = 36), if it exists. If this ratio were to be preserved the atomic weight of thorium (atomic number 90) would be 180, instead of 232, and the thorium nucleus would contain 45 instead of the actual 58 α-particles. However, as the repulsive effect of the net positive charge increases, this relative number of negative electrons becomes too small to overcome the repulsion, and extra groups containing 4 negative to 4 positive electrons, in which this ratio is 1 to 1, are taken up in order to increase the ratio in the nucleus as a whole. The ratio $\frac{\text{Total number of negative electrons}}{\text{Total number of positive electrons}}$ is of fundamental importance in determining in general the stability of complex nuclei,[1] since as the number of α-particles and the resultant positive charge on the nucleus, together with the accompanying repulsion increase, it is necessary to include negative cementing electrons in order to increase the percentage content of negative electricity. *The cementing electrons are thus included in the nuclei of high net positive charge in order to counteract the repulsive effect of the positive charges, by increasing the ratio of negative to positive charges in the nucleus as a whole.* These cementing electrons *add on in*

[1] A recent paper by Kossel considers the ratio $\frac{\text{No. of formula electrons}}{\text{No. of formula } \alpha\text{-particles}}$ in this connection. *Physik. Z.*, 12, 265–9 (1919). This ratio will be considered in a later part of the present paper, under cementing electrons.

pairs, so for each addition the above ratio increases suddenly, and then, as more α-particles add on by themselves, the ratio decreases very slowly, until finally the net positive charge increases sufficiently to make another increment in the ratio essential for the maintenance of stability. According to this hypothesis, if the number of cementing electrons were to be plotted on the Y-axis, and the nuclear charge on the X-axis, a horizontal line coinciding with the X-axis would be obtained until atomic number 18 is reached, when the line would rise to 2 electrons. This is shown in Fig. 2. To the right of atomic number 18 the general form of the cementing electron plot should be given by a second horizontal line (indicated in the figure) extending toward the right, another sudden rise to 2 electrons, etc., that is by a series of horizontal steps of different width, but with a uniform vertical spacing (of 2 electrons). The lowest platform or step is the widest (in the horizontal or N or P direction), and in general the steps become somewhat narrower as the number of cementing electrons increases. The average width is about 2.7 α changes per step after the rise once begins.

The Ratio of Negative to Positive Electrons (N/P) in the Nucleus.

Fig. 3 gives the total number of negative nuclear electrons (N) on the Y-axis, and the number of positive electrons (P) on the X-axis. A slanting straight line drawn upward toward the right, starting at the origin, has a slope equal to $^1/_2$ or 0.5, *the normal slope for light atoms*. On this plot atoms of the helium-thorium series are represented by open circles, the meta-neon-uranium series by circles which are linked in, the lithium series by triangles, and meta-chlorine by a square. A cross indicates that the element weight (ordinary atomic weight) only, is known, and that the element is supposed to be a mixture of isotopes belonging to different series. It may be noted that up to about calcium the helium series atoms lie on this 0.5 line, those of the lithium series very slightly above it, and the meta-neon series still higher. Although meta-magnesium, and meta-silicon, have not been discovered, black circles which represent them, based on the hypothesis that they are members of the meta-neon series, are included in the plot in order to indicate what their relations would be. This is also true in all of the other figures of this general type. While there is little doubt that these meta elements exist, it is possible that they belong to another series. From calcium on, the slope of the plot is seen to be greater than 0.5. It may be seen that most of the elements are represented as, or assumed to be, mixtures. From nickel to uranium the whole plot will undoubtedly divide into a number of series similar to those represented on an enlarged scale at the top of the figure, where the radioactive atoms are plotted. However, between the radioactive atoms and the light atoms, other series than those thus far found among the heavy atoms, such as

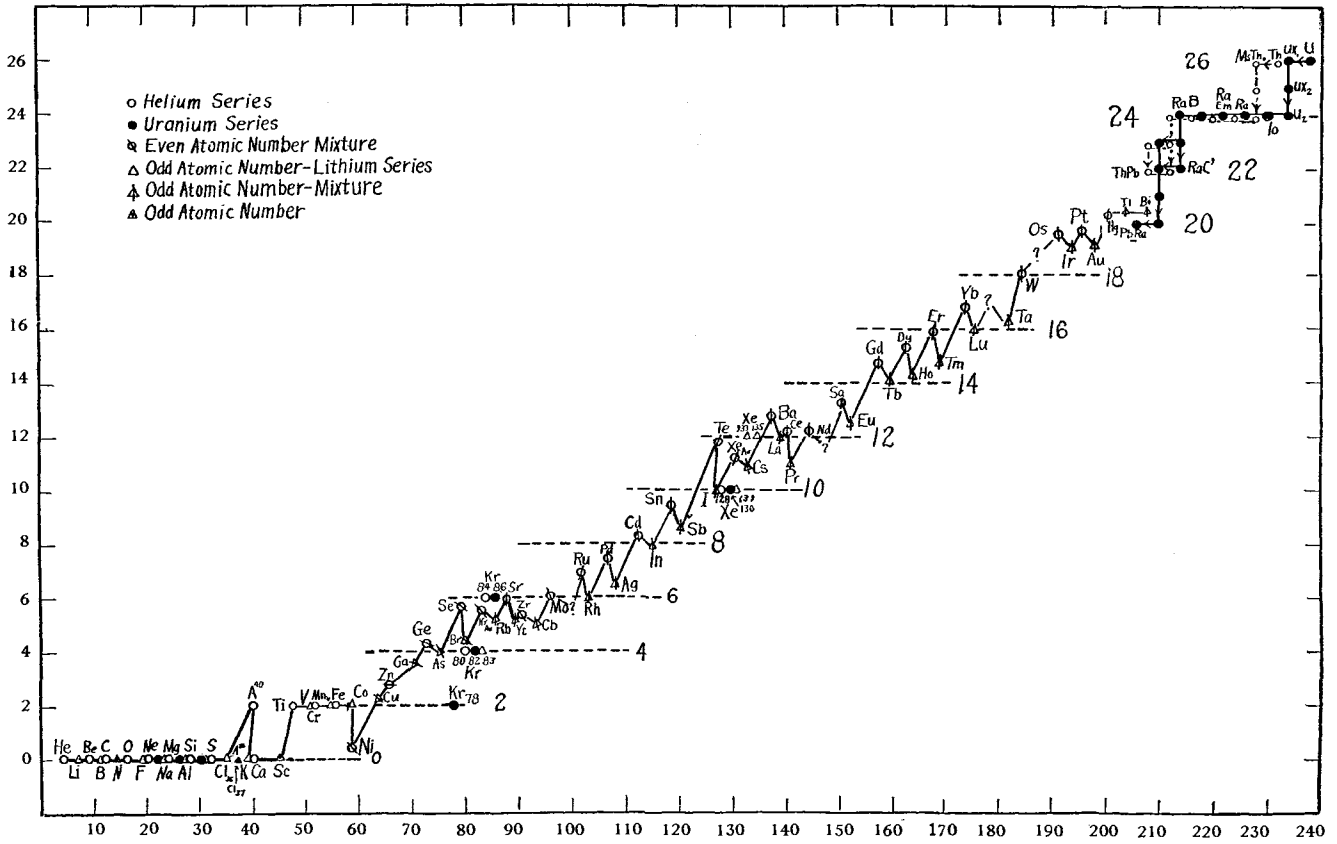

Fig. 2.—Cementing or β-Electrons in the Nuclei of Atoms.

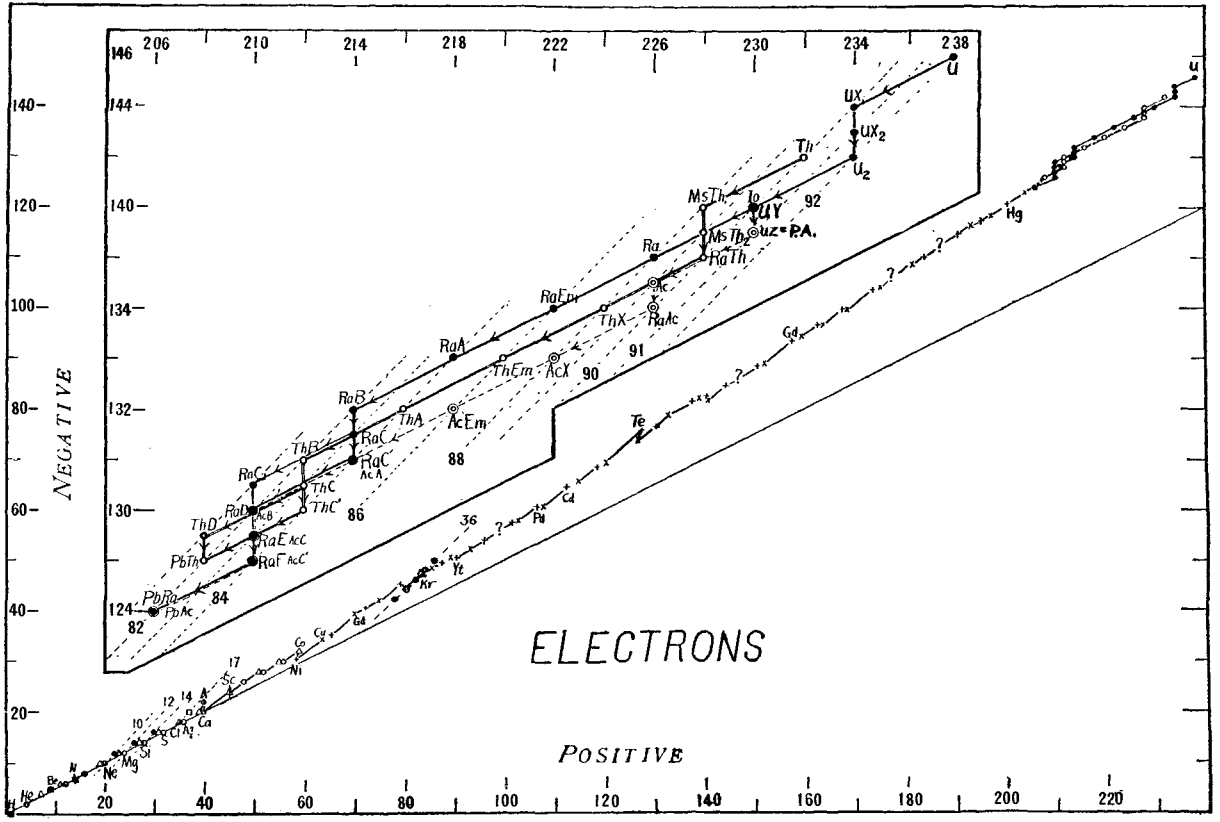

Fig. 3.—Positive and negative electrons in atom nuclei.

(Note: The position of the actinium series is still uncertain. Any later change will be to shift the whole series down or *much more probably up* along the isotopic lines.)

the lithium series, will be undoubtedly found; so from nickel to lead the final plot should be even more complicated than that given for the radio-elements. The dotted lines on the plot have a slope of 1 : 1, or of 45°. Thus they are lines along which the difference $P - N$ is constant, and since $P - N$ gives the nuclear charge or the atomic number, these are isotopic lines, or lines which give the same atomic number. The actinium series is assumed to be derived from uranium II. If it is derived from uranium itself the only change necessary in the diagram is to shift the whole actinium system two places upward along these isotopic lines.

Column *r*, or N/P, in Table I shows that the ratio of negatives to positive electrons for all of the light complex nuclei of even atomic number (sub-column *He*) with the exception of beryllium,[1] is exactly one to two, or 0.5. In the argon nucleus (of at. wt. 40) this ratio suddenly rises to 0.55, falls to 0.5 in calcium, rises suddenly again to 0.542 in titanium, after which it falls gradually to 0.536 in iron. *The subsequent fall to 0.522 in nickel probably indicates that one of the abundant isotopes in nickel has an atomic weight equal to 56*, and that this isotope belongs to the pure α division of the helium series. According to the theory advanced in this paper, the *sudden rise* in the ratio, and its subsequent *gradual fall*, should be characteristic of each series of atoms through its entire course after the first rise takes place, the rise with increasing atomic number being coincident with the addition of *two* cementing electrons, while the gradual fall is due to the addition of α-particles in which this ratio is 0.5, which, it is apparent, has the effect of reducing any ratio higher than itself. Table I (Sections C, D and E) shows that this is exactly what happens in each radio series. Consider, for example, in inverse order of the disintegrations which occur, the primary branch of the uranium series, beginning with radium G, the lightest atomic species. The ratio starts at 0.602, falls to 0.600 (α-addition), rises (β-additions) quickly to 0.605 and 0.610, then falls again by 0.002 to 0.608, rises abruptly to 0.612 and 0.617, and then falls by 4 steps of 0.002 to 0.607, rises to 0.612 and 0.616, and finally ends in 0.614 in uranium. The ratios in the thorium and actinium series follow a similar set of changes, with very nearly the same values.

These relations may be made more prominent by considering not these ratios themselves, but the excess of the ratio over the lowest constant value (0.5), that is $N/P - 0.5$. In the table these numbers have been multiplied by 4 and are given in the Col. 4 ($N/P - 0.5$).

In the lithium series the N/P values start at higher values for the light atoms than in the helium series. Thus it is about 0.52 for fluorine, sodium, and aluminum, rises to about 0.55 for vanadium, manganese and

[1] Leaving out of consideration the less abundant isotopic series of atoms (Sub-Column *U*).

cobalt, which is not much higher than in the helium series. Finally, after nickel is passed, and the mean number of cementing electrons begins to rise steadily, the ratio falls to practically the same values as, and even below those for the helium series, but shows the same gradual rise, reaching the value 0.602 in iridium. The ratio is specially high in both of the isotopic (meta-neon and meta-chlorine) series, being about 0.54 for meta-neon (meta-magnesium, meta-silicon) and meta-chlorine. This particularly high ratio is due to the presence of the μ group, so it is found on comparing members of the uranium and thorium series containing the same number of α-particles, that the ratio is always higher in the uranium derivative. The ratio is specially high (0.572) in lithium, and also high in beryllium (0.556), and boron (0.545).

The Abundance of Atomic Species as Related to the Ratio (N/P) of Negative to Positive Electrons in the Nucleus.

The details of the abundance relations of the elements in the crust of the earth and the meteorites have been treated in an earlier paper of this series.[1] While the ratio N/P is not the only factor upon which the stability of an atom depends, it is evidently of primary importance. *Thus lithium, beryllium, and boron, with the relatively high values of the ratio given in the preceding paragraph, are rare both in the metorites and on earth.* Carbon, the first atom in the system in which the ratio falls to the normal value 0.5, is the lightest complex atom found in any considerable quantity in the meteorites, and is about 26 times more abundant in the lithosphere than any of the three atomic species just mentioned. Nitrogen, which also has a ratio of 0.5, is, however, not abundant, indicating that other factors are of importance.[2] However, among the elements of atomic number less than 18, all of the abundant atomic species, oxygen, magnesium, silicon, and sulfur, have just this value of the ratio. Iron, in whose nuclei this ratio has a value 0.536, is, nevertheless, a very abundant element, but its nuclear charge is somewhat high (26) which would increase the value of N/P essential for stability, according to the theory presented earlier in this paper.

Both from the known data on the abundance of the elements in the meteorites, and from Clarke's estimate of the composition of the crust of the earth, *it is evident that no atomic species which has a ratio of N/P greater than 0.54 (or a net positive nuclear charge higher than 29) occurs in an atomic percentage greater than 0.01.* In other words, all such elements and atomic species are relatively very rare.

[1] "The Evolution of the Elements and the Stability of Complex Atoms," THIS JOURNAL, 39, 856–79 (1917).

[2] The nitrogen nucleus contains an *odd number of negative electrons.* In an earlier paper it has been pointed out that the presence of an odd number of negative electrons is always accompanied by a low stability for a radioactive atom, and a low abundance for other atoms, which also indicates a low stability.

Cementing and Excess Electrons in Atom Nuclei.

The early papers of this series gave 2 equations[1] for the atomic weights of the various atomic species, and from them calculated the number of cementing electrons (c) in the light[2] and also in the heavy[3] atoms. If W is taken to represent the atomic weight, then

$$W = P \qquad (1)$$

where P is the number of positive electrons in the nucleus of each atom if the weight is that for a pure species of atoms, but is merely an average value if the elements is a mixture and the element weight is used. Now for atoms of the *helium-thorium series:*

$$P = 2(n + c) = W \qquad (2)$$

where n is defined by the equation

$$n = P - N \qquad (3)$$

in which N represents the total number of negative electrons in the nucleus. Thus n is the net positive charge on the nucleus, which is equal to the atomic number, and also to the number of non-nuclear or planetary electrons. It is evident that for the calculation of the number of α-cementing electrons, Equation 2 may be put in the following form,

$$c = W/2 - n = P/2 - n = \frac{P - 2n}{2} \qquad (4)$$

That is, the number of cementing electrons is equal to the excess of half the number of positive electrons over the net positive nuclear charge. The latter is sometimes called the number of "free" positive electrons in the nucleus, but there is considerable danger that this term may be interpreted in a false sense, since all of the positive electrons in the α-particles present are bound with exceeding firmness.

The above formulas are valid for *only* the members of the *helium-thorium series*, but they may be used for the *lithium series also, provided* an extra term is added. This addition gives

$$P = W = 2(n + c) + 1/2 + 1/2(-1)^{n-1} \qquad (5)$$

$$c = W/2 - n - 1/4 - 1/4\,(-1)^{n-1} \qquad (6)$$

While Equations 5 and 6 hold for both the helium and lithium series, they are not valid for *metaneon-uranium series*. For this last series the following relation is true

$$c = W/2 - n - 1 = P/2 - n - 1. \qquad (7)$$

While it might be possible to combine Equation 7 with Equation 6 in such a way as to give one equation for the number of cementing electrons

[1] THIS JOURNAL, **37**, 1380, 1385, 1386, 1395 (1915).
[2] *Ibid.*, **39**, 857–9 (1917).
[3] *Phys. Rev.*, **15**, 78–79, 77, 85–94 (1920).

in all 3 series, the resulting formula would be so complicated that it is more convenient to use the equations separately.

In 1917 Durrant[1] made a study of our Equations 2 to 5, and in agreement with the structural formulas for the atoms published one month later by the writer, showed that c remains zero up to argon, then suddenly increases by 2, decreases to 0 in potassium, calcium, and scandium, increases again by 2, and remains at that value up to atomic number 27 (cobalt). Durrant plotted c and showed that at atomic number 29 its value begins to increase with an almost constant slope equal to $1/3$, but that this slope increases slightly when the radioactive elements are reached. He supported the idea presented in the first paper of this series, according to which this is a region of numerous isotopes and possibly consists partly of the remnants of radioactive series, which may still be disintegrating with extreme slowness, and may extend downward as far or farther than iron.

Whenever Equations 2 and 4 are used for members of the uranium or lithium series, they should be represented in a slightly different way in order to prevent confusion.

$$W = 2(n + f) = P \qquad (8)$$

or

$$f = W/2 - n \qquad (9)$$

where f is a function whose values are equal to the number of cementing electrons whenever the atomic species is a member of the helium-thorium series, but *does not represent any specific electrons when it is applied to any other series, since it indicates the presence of an odd number of cementing electrons in atoms of the uranium series when an even number are present, and the presence of half of a cementing electron in many of the atoms of the lithium series.* In spite of this fact Kossel,[2] in 1919, plotted the values of f as obtained from our Equation 9, and considered the equation to give the number of cementing, or of what he calls beta (β) electrons, in the uranium as well as in the thorium series. The plot which he obtained is practically the same as that obtained several years earlier by Durrant.

A Theoretical Basis for the Rule of Fajans.

The rule of Fajans states that when isotopes alone are considered it is found that the β-disintegrations *increase* in velocity and the α disintegrations *decrease* in velocity, as the atomic weight rises. In order to give this rule greater theoretical significance Kossel puts it is essentially the following form: The greater the ratio of the number of β (cementing) electrons (β) to the number of α-particles (α) in the nucleus, that is the

[1] Durrant, THIS JOURNAL, 39, 621–7 (1917).

[2] Kossel, *Physik. Z.*, 20, 2659 (1919). The papers of Durrant and of Kossel should be consulted in the original, together with the earlier papers by the writer, if a proper view of their interrelations is to be obtained.

greater β/α, the more rapid is the β disintegration which lowers the ratio, and the less rapid is the α disintegration which raises it. Unfortunately, this formulation of the rule, while extremely suggestive, is nevertheless incorrect. However, Kossel also states the rule in another form, in which he replaces the ratio β/α as given above, by the ratio f/W. In this second form the rule is found to hold as well as the Fajans rule from which it is derived, though it will be shown later that it is not entirely exact, since when the differences in the values of the ratio f/W are too small, certain specific factors sometimes obscure the relationship.

The explanation of the above discrepancy lies in the fact that Kossel considers that the ratios β/α and f/W are proportional to each other, which is not the case, as may be easily seen. Thus since

$$P = W \quad f/W = f/P. \tag{10}$$

From (3), (8) and (10)

$$f/P = P/2P - N/P = 1/2 - \frac{P - N}{P} = N/P - 1/2. \tag{11}$$

Now $P/4$ is *approximately* equal to the number of α-particles in the nucleus, and f is *approximately* equal to the number of cementing electrons, so

$$4f/P = 4(N/P - 1/2) \tag{12}$$

is approximately equal to the ratio β/α, but the approximation is not close enough to make the Kossel relation valid when expressed in terms of the latter ratio.

The equations given above indicate that the real theoretical basis of the rule of Fajans does not lie in the ratio β/α, but in the term $N/P - 0.5$, which is the excess of the ratio of total nuclear negative to positive electrons, over the normal value (0.5) of N/P in helium itself, and in the light atoms of the helium series. Since the relation holds for $N/P - 0.5$, it must also hold for the values of N/P, which is the important factor, and not the values of β/α.

The rule of Fajans may now be stated in the following new form. *The greater the ratio* $\frac{\text{Total number of negative electrons}}{\text{Total number of positive electrons}}$ *in the nucleus the more rapid is a β-disintegration which lowers the ratio, and the less rapid is an α-disintegration which raises it, provided the net positive charge on all of the nuclei considered is the same,* that is, when isotopes alone are considered.[1] In Fig. 3 the ratio N/P increases with the height along an isotopic

[1] *Note on a Reversal of the Above Relationship.* The general idea on which the above relationship is based, is that as it becomes more negative in the sense that N/P increases, the nucleus binds positively charged particles more firmly, and negatively charged particles less firmly. While if isotopic atoms alone are considered, that is nuclei of constant net charge, there are only two complete exceptions to the relationship in about 37 disintegrations, there is an apparent reversal of the rule when the

line, and, therefore, the period of β disintegrations decreases, and of the α disintegration increases, with the height along these lines. This figure gives a plot which may be considered to present the normal or equilibrium values of N and P for the more stable types of atoms. It will be seen that while *the isotopes of krypton*[1] *differ in atomic weight by 8, a considerable difference, none of these isotopes lies very far away from the line representing the mean values of N and P for the adjacent elements.* Aston's work indicates that Kr_{78}, the isotope which lies farthest away from the line of mean values, is present in krypton in only very small quantities. Figs. 4 and 5 give the values of the function f, that is of $N/P - 1/2$, so they represent the rise of the N, P plot in Fig. 3 above the straight line which has the normal slope equal to $1/2$. The *heavy* line in Fig. 5 represents the number of *cementing electrons*, given separately in Fig. 2, while the lighter lines plot the values of the function f. It will be noted that both Fig. 4 and Fig. 5 have the same general form, and that the former appears very much as if it were the latter plot, the peaks of which have all been pushed over toward the right. This is due to the fact that the higher the f values in isotopic species of atoms ($P - N$ = const.), the higher is the total number of negative electrons, and therefore the greater is the value of P, which is plotted on the X-axis in Fig. 4.

In the principal plots of these figures an open circle represents a member of the helium-thorium series of atoms between atomic numbers 2 nuclear charge varies. There are also 3 partial exceptions to the rule, in that members of the radium and actinium series of the same N/P ratio, have different periods, but this is to be expected as the result of more specific influences. These statements are based on the idea that the actinium series springs from U_{II}. Thus the reversal occurs in successive α disintegrations, since as is well known, the period and, therefore, the stability of the nucleus, with reference to the giving off of α-particles, decreases rapidly as the disintegration proceeds. That is, as it becomes *less* positive, both with respect to its net charge and with reference to the ratio N/P, the nucleus holds each succeeding positive particle *less* firmly, so as the relative negative electron content increases, the stability with reference to the retention of positive α-particles decreases. In the radium series there are 5, and in both the actinium and the thorium series as now known, 4 such successive α-disintegrations. There is also a reversal with respect to the loss of negative electrons, since the second β-disintegration is always more rapid than the first in the case of 2 successive changes of this nature, even though the nucleus is becoming more positive by the loss of its β-particles. There is thus an irregular, but periodic variation in the stability of nuclei in any one series. As an hypothesis to account for this fact it has been suggested in an earlier paper that the negative cementing electrons are grouped in pairs, and that in the nucleus the α-particles are grouped in aggregates containing 4 or 5 (and probably other numbers, as low as one) such particles in each group, and that each aggregate becomes less stable as it decreases in size. (*Phys. Rev.*, 15, 88 (1920)).

[1] When this paper was sent to THIS JOURNAL Aston's note in *Nature*, giving news of his work on the isotopes of krypton, xenon, etc., had not been received. The discussion presented here, and the values representing the isotopes of silicon as presented in Table I and Fig. 4, were inserted later by permission of the editor.

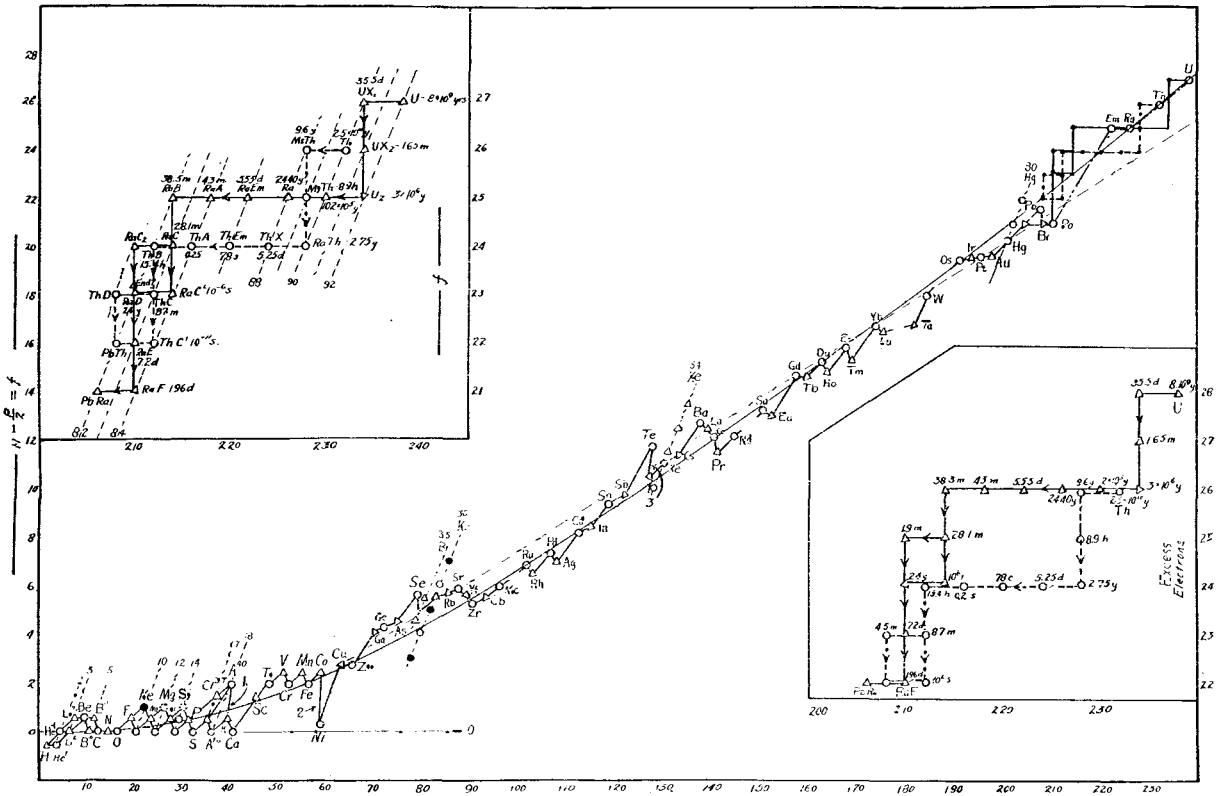

Fig. 4.—(NOTE: This figure was revised after the completion of the paper in order to include new works by Rutherford and by Aston.)

(The ordinates represent $f = N - P/2$. The small letter n given above should be replaced by N.)

and 27, but from 28 to 80 they represent only that the atomic number is even, except where more than one isotope is given. With the same conventions, an open triangle represents either a lithium series atom, or an element of odd atomic number, while an inked in circle indicates a number of the metaneon-uranium series, and a similar triangle, of the metachlorine series.

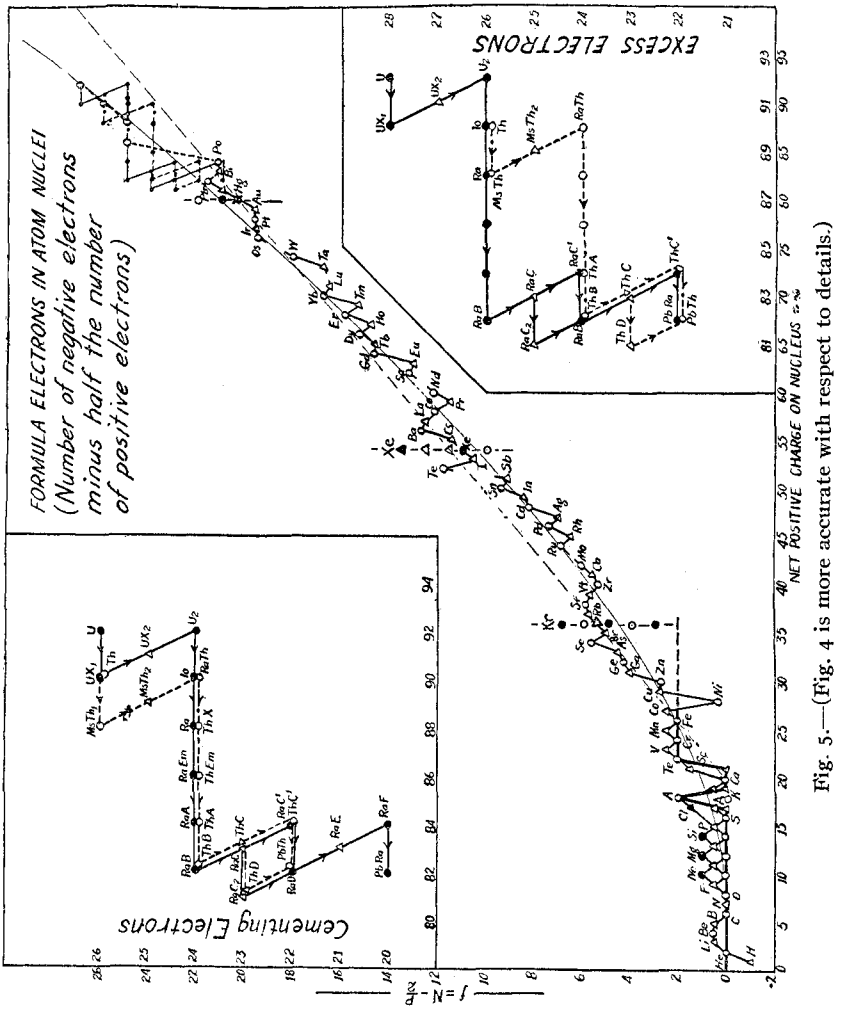

Fig. 5.—(Fig. 4 is more accurate with respect to details.)

It will be seen that Kr_{80} in Fig. 3 is almost coincident with $Br_{av.}$ and Kr_{86} is not far from $Sr_{av.}$. The number of formula electrons in krypton is 3, 4, 5, $5^1/_2$, 6 and 7 for the atomic weights 78, 80, 82, 83, 84 and 86. Aston finds the atomic weights of xenon to be 128, 130, 131, 133 and 135, and this indicates formula electron contents 10, 11, $11^1/_2$, $12^1/_2$, and $13^1/_2$.

TABLE I.—POSITIVE AND NEGATIVE ELECTRONS IN THE NUCLEI OF ATOMS.

n. Net positive charge.	*b.* Element or atomic species.	*P.* Positive electrons.	α-Particles.	*N.* Negative electrons.	*r.* Ratio *N/P.* He.	U.	*o.*	*C.* Cementing electrons.	*E.* Excess electrons. He.	U.	*o.*	*f.* Formula electrons. He.	U.	*o.*	$4(N/P - 1/2)$. He.	U.	*o.*
1	Hydrogen	1		0										0.5			
2	Helium	4	1	2	0.5							0.0			0.0		
3	Lithium	6		3			(0.5)										
3	Lithium	7	1	4			0.572	0			2			0.5			0.288
4	Beryllium	9	2	5	0.555			0	(1)			(0.5)			(0.22)		
5	Boron	10	2	5			(0.5)	0									
5	Boron	11	2	6			0.545	0			2			0.5			0.180
6	Carbon	12	3	6	0.5			0	0			0.0			0.0		
7	Nitrogen	14	3	7			0.5	0			1			0.0			0.0
8	Oxygen	16	4	8	0.5			0	0			0.0			0.0		
9	Fluorine	19	4	10			0.526	0			2			0.5			0.104
10	Neon	20	5	10	0.5			0	0			0.0			0.0		
	Meta-neon	22	5	12		0.545		0		2			1.0			0.180	
11	Sodium	23	5	12			0.522	0			2			0.5			0.088
12	Magnesium	24	6	12	0.5			0	0			0.0			0.0		
	Meta-magnesium	26	6	14		0.538_5		0		2			1.0			0.144	
13	Aluminum	27	6	14			0.518_6	0			2			0.5			0.074

14	Silicon	28	7	14	0.5			0	0		0.0			0.0		
	Silicon 29	29	7	15	(0.517)			0								
	Meta-silicon	30	7	16		0.5333		0		2		1.0			0.133	
15	Phosphorus	31	7	16			0.516	0		2		0.5				0.064
16	Sulfur	32	8	16	0.5			0	0		0.0			0.0		
17	Chlorine	35	8	18			0.5145	0		2		0.5				0.058
	Meta-chlorine	37	8	20		0.541		0		4		1.5			0.164	
	Chlorine 39	39	9	22			0.564	2								
18	Argon 36	36	9	18	0.5			0								
18	Argon	40	10	22	0.550			2	2		2.0			0.200		
19	Potassium	39	9	20			0.513	0		2		0.5				0.052
20	Calcium	40	10	20	0.5			0	0		0.0			0.0		
21	Scandium	45	11	24			0.5335	0		2		1.5				0.134
22	Titanium	48	12	26	0.542			2	2		2.0			0.168		
23	Vanadium	51	12	28			0.549	2		4		2.5				0.196
24	Chromium	52	13	28	0.539			2	2		2.0			0.156		
25	Manganese	55	13	30			0.546	2		4		2.5				0.184
26	Iron 52	52	13	26	0.5			0	0		0			0		
26	Iron	56	14	30	0.536			2	2		2.0			0.144		
27	Cobalt	59	14	32			0.543	2		4		2.5				0.172

TABLE I, B.—HEAVY ATOMS (AVERAGE VALUES FOR MIXTURES OF ISOTOPES).

n. Net positive charge.	*b.* Element or atomic species.	*P.* Positive electrons.	α-Particles.	*N.* Negative electrons.	*r.* Ratio N/P. *e.*	*u.*	*o.*	*f.* Formula electrons. *e.*	*u.*	*o.*	$4(N/P - 1/2)$. He.	*u.*	*o.*
28	Nickel	58.68	14	30.68	0.522$_5$			1.34			0.090		
29	Copper	63.57	15	34.57			0.544			2.78			0.176
30	Zinc	65.37	16	35.37	0.541			2.68			0.164		
31	Gallium	70.1	17	39.1			0.558			4.05			0.232
32	Germanium	72.5	18	40.5	0.555			4.25			0.220		
33	Arsenic	75	18	42			0.560			4.48			0.240
34	Selenium	79.2	19	45.2	0.570$_5$			5.6			0.282		
35	Bromine	79.92	20	44.92			0.562			4.96			0.248
36	Krypton	82.92	20	46.92	0.566			5.46			0.264		
37	Rubidium	85.45	21	48.45			0.567			5.72			0.268
38	Strontium	87.63	21	49.63	0.566$_5$			5.81			0.266		
39	Yttrium	89.33	22	50.33			0.563$_5$			5.66			0.254
40	Zirconium	90.6	22	50.6	0.558$_5$			5.3			0.234		
41	Columbium	93.1	23	52.1			0.559$_5$			5.55			0.238
42	Molybdenum	96.0	24	54	0.562$_5$			6.0			0.250		
43	Eka-manganese												
44	Ruthenium	101.7	25	57.7	0.577$_5$			6.85			0.310		
45	Rhodium	102.9	25	57.9			0.562$_5$			6.45			0.250
46	Palladium	106.7	26	60.7	0.569			7.35			0.2756		
47	Silver	107.88	26	60.88			0.564$_5$			6.94			0.2576
48	Cadmium	112.40	28	64.40	0.573			8.2			0.2920		
49	Indium	114.8	28	65.8			0.573			8.4			0.2928
50	Tin	118.7	29	68.7	0.578			9.35			0.3150		
51	Antimony	120.2	30	69.2			0.576			9.1			0.30218
52	Tellurium	127.5	31	75.5	0.592			11.75			0.36817		
53	Iodine	126.92	31	73.92			0.582			10.46			0.32918
54	Xenon	130.2	32	76.2	0.585			11.1			0.341		
55	Cesium	132.81	33	78.81			0.594			11.40			0.344
56	Barium	137.37	34	81.37	0.592			12.68			0.369		
57	Lanthanum	139.0	34	82.0			0.590			12.5			0.360

n.		P.	α.	N.	e.	o.			
58	Cerium	140.25	35	82.25	0.586		12.12		0.346
59	Praseodymium	140.9	35	81.9		0.581		11.45	0.325
60	Neodymium	144.3	36	84.3	0.584		12.15		0.337
61	Eka-neodymium								
62	Samarium	150.4	37	88.4	0.588		13.2		0.351
63	Europium	152.0	38	89.0		0.586		13.00	0.342
64	Gadolinium	157.3	39	93.3	0.593		14.63		0.372
65	Terbium	159.2	39	94.2		0.592		14.6	0.367
66	Dysprosium	162.5	40	96.5	0.594		15.25		0.375
67	Holmium	163.5	40	96.5		0.590		14.75	0.361
68	Erbium	167.7	41	99.7	0.594		15.85		0.378
69	Thulium	168.5	42	99.5		0.590		15.25	0.362
70	Ytterbium	173.5	43	103.5	0.596_5		16.75		0.386
71	Lutecium	175.0	43	104.0		0.594_5		16.50	0.377
72	Eka-lutecium								
73	Tantalum	181.5	45	108.5		0.598		16.75	0.369
74	Tungsten	184.0	46	110.0	0.598		18.0		0.391
75	Eka-manganese II								
76	Osmium	190.9	47	114.9	0.602		19.45		0.407_5
77	Iridium	193.1	48	116.1		0.602—		19.55	0.405
78	Platinum	195.2	48	117.2	0.601—		19.6		0.401
79	Gold	197.2	49	118.2		0.600		19.6	0.397
80	Mercury	200.6	50	120.6	0.602		20.3		0.405_5
81	Thallium	204.0	51	123.0		0.603		21.0	0.412—
82	Lead	207.2	51	125.2	0.604		21.6		0.416_5
83	Bismuth	208.0	52	125.0		0.601		21.0	0.404
84	Polonium	210.0	52	126.0	0.600		21.0		0.400
85	Eka-iodine		55						
86	Radium emanation	222.0		136.0	0.613		25.0		0.450
87	Eka-cesium								
88	Radium	226.0	56	138.0	0.611		27.0		0.478
89	Actinium								
90	Thorium	232.15	58	142.15	0.612		26.0		0.448
91	Protoactinium	234		143		0.612—		26.0	0.445—
92	Uranium	238.16	59	146.16	0.614		27.0		3_5 50.4

TABLE I, C.

Uranium Series in Order of Disintegration.

Element.	Net positive charge.	Atom.	Positive electrons.	Negative electrons.	α-Particles.	N/P.	Electrons. Cementing.	Excess.	Formula.	$4(N/P - 0.5)$.
U	92	U	238	146	59	0.614	26	28	27	0.4538
Th	90	UX_1	234	*144*	*58*	*0.616*	*26*	*28*	*27*	*0.4615*
Bv	91	UX_2	234	*143*	*58*	*0.612—*	*25*	*27*	*26*	*0.4446*
U	92	U_2	234	142	58	0.607	24	26	25	0.4273
Th	90	Io	230	140	57	0.609	24	26	25	0.4350
Ra	88	Ra	226	138	56	0.611	24	26	25	0.4425
Nt	86	RaEm	222	136	55	0.613	24	26	25	0.4505
Po	84	RaA	218	134	54	0.615	24	26	25	0.4587
Pb	82	RaB	214	*132*	*53*	*0.617*	*24*	*26*	*25*	*0.4672*
Bi	83	RaC	214	*131*	*53*	*0.612*	*23*	*25*	*24*	*0.4482*
Po	84	RaC'	214	130	53	0.608	22	24	23	0.4300
Pb	82	RaD	210	*128*	*52*	*0.610*	*22*	*24*	*23*	*0.4380*
Bi	83	RaE	210	*127*	*52*	*0.605*	*21*	*23*	*22*	*0.4190*
Po	84	RaF	210	126	52	0.600	20	22	21	0.4000
Pb	82	RaG	206	124	51	0.602	20	22	21	0.4078
			Secondary Branch of Uranium Series.							
Pb	82	RaB	214	*132*	*53*	*0.617*	*24*	*26*	*25*	*0.4672*
Bi	83	RaC	214	131	53	0.612	23	25	24	0.4482
Tl	81	RaC_2	210	*129*	*52*	*0.614_3*	*23*	*25*	*24*	*0.4570*
Pb	82	End	210	128	52	0.609_5	22	24	23	0.4380

TABLE I, D.

Thorium Series in Order of Disintegration.

Th	90	Th	232	142	58	0.612	26	26	26	0.4485
Ra	88	$MsTh_1$	228	*140*	*57*	*0.614*	*26*	*26*	*26*	*0.4562*
Ac	89	$MsTh_2$	228	*139*	*57*	*0.610*	*25*	*25*	*25*	*0.4387*
Th	90	RaTh	228	138	57	0.605	24	24	24	0.4210
Ra	88	ThX	224	136	56	0.607_1	24	24	24	0.4285
Nt	86	ThEm	220	134	55	0.609_1	24	24	24	0.4363
Po	84	ThA	216	132	54	0.611_1	24	24	24	0.4445
Pb	82	ThB	212	*130*	*53*	*0.613_2*	*24*	*24*	*24*	*0.4527*
Bi	83	ThC	212	*129*	*53*	*0.608_8*	*23*	*23*	*23*	*0.4343*
Po	84	ThC'	212	128	53	0.603_8	22	22	22	0.4150
Pb	82	PbTh	208	126	52	0.606	22	22	22	0.4230
			Secondary Branch of Thorium Series.							
Pb	82	ThB	212	*130*	*53*	*0.613_2*	*24*	*24*	*24*	*0.4527*
Bi	83	ThC	212	129	53	0.608_6	23	23	23	0.4343
Tl	81	ThD	208	*127*	*52*	*0.610_5*	*23*	*23*	*23*	*0.4421*
Pb	82	End?	208	126	52	0.606—	22	22	22	0.4230

TABLE I, E.

Actinium Series (Assumed to be Derived from U[2]).[a]

											$4(N/P-1/2)$ if from U.
Th	90	UY	230	140	57	0.608_8	24	26	25	0.4350	0.4615
Pa	91	Eka-Ta	230	139	57	0.604_5	23	25	24	0.4172	0.4446
Ac	89	Ac	*226*	*137*	*56*	0.606_2	*23*	*25*	*24*	*0.4248*	*0.4523*
Th	90	RaAc	226	136	56	0.602—	22	24	23	0.4070	0.4350
Ra	88	AcX	222	134	55	0.603_6	22	24	23	0.4144	0.4425
Nt	86	AcEm	218	132	54	0.605^4	22	24	23	0.4220	0.4505
Po	84	AcA	214	130	53	0.607_5	22	24	23	0.4298	0.4587
Pb	82	AcB	*210*	*128*	*52*	0.609_5	*22*	*24*	*23*	*0.4380*	*0.4672*
Bi	83	AcC	210	127	52	0.605	21	23	22	0.4192	0.4482
Po	84	AcC′	210	126	52	0.600	20	22	21	0.4400	0.4300
PbAc	82	AcE	206	124	51	0.602	20	22	21	0.4078	0.4380
Pb	82	AcD	206	124	51	0.602	20	22	21	0.4078	..

P = no. of positive electrons, N = no. of negative electrons.

[a] E. Q. Adams considers that the atomic weight of lead derived from actinium is 207. If this is the case all of the values for the actinium series should be recalculated.

The cementing electron content is of much more interest (Fig. 2). For the lowest krypton it is 2, for the next 3 higher it is 4, and for the 2 highest it is 6, so if the heavy cementing electron line were to be extended toward the right in Fig. 5, there would be 3 horizontal steps at 2, 4 and 6. This indicates, too, that the step for 2 cementing electrons, extends from argon to krypton, or from atomic number 18 to 36 at least, which is almost equal to the range of the 0 step from helium to calcium. However, as has already been stated, it is extremely likely that one of the isotopes of nickel (No. 28) has a zero cementing electron content, so the extent of the zero step is considerable (equal to about 26). The number of cementing electrons in the lowest 3 xenons is 10, in the next higher is 12, and in the highest it is uncertain. If this xenon 135 is a member of the meta-chlorine series, it contains 12 such electrons and there are only 2 such steps. Here the cementing electrons are only those concerned in cementing on extra α-particles. Whether electrons may be given off from ν or μ groups has not been determined, if not, such electrons should be classed as binding electrons.

Col. E, in Table I, gives the number of *excess* electrons e where this term indicates all of the nuclear negative electrons which are not contained in the α-particles themselves, or

$$e = N - N_{\alpha b} \tag{13}$$

where $N_{\alpha b}$ is the number of *binding* electrons in the α-particles.

In the helium-thorium series

$$e = c \tag{14}$$

in the metaneon-uranium series

$$e = W/2 - n + 1 = P/2 - n + 1 \tag{15}$$

and in the lithium series

$$e = c + 2 = P/2 - n + 3/2. \qquad (16)$$

In Table I the sub-columns marked He include members of the helium-thorium series, those marked U include members of the metaneon-uranium series, *o* indicates that the sub-column relates to atoms of odd, and *e* that it relates to atoms of even atomic number.

Suggested Formulas for the More Abundant Light Atoms.

Formulas for the light atoms which are in agreement with the data of Table I, are given in Table II. These formulas were given in part in an earlier paper in THIS JOURNAL, but it is desirable to reproduce them to indicate minor changes and to correct errors in typesetting.

TABLE II.

Hypothetical Composition of Light Atoms and their Nuclei.

	Formula of nucleus.						Non-nuclear electrons.		
			Lithium series.		Metachlorine series.				
Symbol.	Helium series.	Metaneon series.	*a.*	*b.*	*a.*	*b.*	Inner.	Middle.	Valence or outer.
He.....	α	..	...	...	...	...	e_2	...	..
Li.....	..	..	λ	$\alpha\nu$	...	...	e_2	...	e
Be.....	$(\alpha_2\eta\beta)$ or $(\eta_3\beta)_3\beta_2$	..	...	...	...	...	e_2	...	e_2
B.....	..	..	$\lambda\alpha$	$\alpha_2\nu$	...	...	e_2	...	e_3
C.....	α_3	..	...	...	...	...	e_2	...	e_4
N.....	..	..	...	$(\alpha_3\mu_2\beta)$	...	...	e_2	...	e_5
O.....	α_4	..		...	...	...	e_2	...	e_6
E.....	..	..	$\lambda\alpha_3$	$\alpha_4\nu$	...	...	e_2	...	e_7
Ne_{20}.....	α_5	..	...	...	...	...	e_2	e_8	..
Ne_{22}.....	..	$\alpha_5\mu$	...	...	...	...	e_2	e_8	..
Na.....	..	..	$\lambda\alpha_4$	$\alpha_5\nu$	...	...	e_2	e_8	e
Mg_{24}.....	α^6	..	...	...	...	...	e_2	e_8	e_2
Mg_{28}.....	..	$(\alpha_6\mu)$	...	...	...	...	e_2	e_8	e_2
Al.....	..	..	$\lambda\alpha_5$	$\alpha_6\nu$	...	...	e_2	e_8	e_3
Si_{28}.....	α_7	..	...	...	...	...	e_2	e_8	e_4
Si_{30}.....	..	$(\alpha_7\mu)$	...	...	...	...	e_2	e_8	e_4
P.....	..	..	$\lambda\alpha_6$	$\alpha_7\nu$	...	...	e_2	e_8	e_6
S.....	α_8	..	...	...	...	...	e_2	e_8	e_5
Cl_{35}.....	..	..	$\lambda\alpha_7$	$\alpha_8\nu$	...	...	e_2	e_8	e_7
Cl_{37}.....	..	..	...	...	$\lambda\alpha_7\mu$	$\alpha_8\nu\mu$	e_2	e_8	e_7
A_{36}.....	α_9	..	...	...	...	...	e_2	$e_8 + e_8$	..
A_{40}.....	$\alpha_{10}\beta_2$	..	...	...	...	...	e_2	$e_8 + e_8$	..
K.....	..	..	$\lambda\alpha_8$	$\alpha_9\nu$	...	...	e_2	$e_8 + e_8$	e
Ca.....	α_{10}	..	...	...	...	...	e_2	$e_8 + e_8$	e_2
Sc.....	..	..	...	$(\alpha_{11}\eta_2\beta)$	...	...	e_2	$e_8 + e_8$	e_3
Ti.....	$\alpha_{12}\beta_2$	..	...	...	...	...	e_2	$e_8 + e_8$	e_4
V.....	..	..	$\lambda\alpha_{11}\beta_2$	$\alpha_{12}\nu\beta_2$	...	...	e_2	$e_8 + e_8$	e_5
Cr.....	$\alpha_{13}\beta_2$	..	...	...	...	...	e_2	$e_8 + e_8$	e_6
Mn.....	..	..	$\lambda\alpha_{12}\beta_2$	$\alpha_{13}\nu\beta_2$	...	...	e_2	$e_8 + e_8$	e_7
Fe.....	$\alpha_{14}\beta_2$	..	...	...	...	...	e_2	$e_8 + e_8$	e_8
Co.....	..	..	$\lambda\alpha_{13}\beta_2$	$\alpha_{14}\nu\beta_2$	...	...	e_2	$e_8 + e_8$	e_9

Columns *a* and *b* give alternative formulas for the respective nuclei, the formulas in Col. *b* being based on the hypothesis that the nucleus of the lithium atom contains one α-particle.

The parentheses around the nuclear formulas for nitrogen and scandium indicate that these nuclei do not belong to the series under which they are placed, while those around the formulas for metamagnesium and metasilicon indicate that these species have not been discovered.

Formulas for the heavy atoms will be found in an earlier paper [1] It should be noted that the formulas given above for A_{40} and Ca, indicate that their atoms are *isomeric*.

The New Periodic System and the Values of $N - \frac{P}{2}$, (equal to $\frac{P}{2} - n$) or the Number of Formula Electrons.

The writer has already presented evidence that there is a periodic variation in the abundance of the elements as a function of the atomic number, and N. F. Hall[2] has shown the same periodicity in the properties of the radioactive elements. Figs. 4 and 5 indicate that there is also a periodic variation of the same general nature, in the values of f, that is in $N - P/2$. Leaving the isotopic atoms out of account, it is apparent that the values of f for the atoms of odd nuclear charge are higher than for those of even number among the atoms from atomic number 8 to 28, that is the peaks occur on the odd numbers. Strangely enough, this relationship is reversed in the average values plotted for elements 31 to 82, for in this range practically all of the peaks occur on the even numbers, and practically all of the troughs on the odd numbers.

The seeming reversal of this relationship seems to be complete in certain ranges. Thus from atomic numbers 44 to 51, and from 59 to 71 (with the possible exception of 61, which has not been discovered) each even number is represented by a peak and each odd number by a trough. If the diagram is examined between atomic numbers 9 and 15 it will be seen that this is, however, just the relationship which also holds in this range if the thorium series atoms are left out of account, and if the uranium or isotopic species of atoms are the only ones considered for the even numbers. Thus the general relations between atomic numbers 44 and 51, and 59 and 71, is just what would be expected if most of the even atomic species in these ranges belong to the uranium, and not to the thorium series, that is if the most abundant isotopes are members of the former series. It is apparent from the large scale plot that in the ranges above specified the mean elevation of the points which represent even, above those which represent the adjacent odd atomic numbers is of about the same magnitude as is found between the uranium and lithium series

[1] *Phys. Rev.*, **15**, 86–89 (1920).

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

among the light atoms. However, that this apparent explanation may not be correct, is indicated by the fact that the same relation is found between thallium (odd), ordinary lead (even), and bismuth. Now it seems probable that ordinary lead is a mixture in almost equal percentages of lead from uranium, so it is possible that the explanation may be found in the electron content of the atoms of odd number.

It is evident that all of the atoms of extremely high relative negative electron content, have even atomic numbers. For example, tellurium (52) particularly, and barium (56), and selenium (34), lie on especially high peaks. The atoms from 31 to 34, and much more markedly the radioactive atoms from radium to uranium, exhibit abnormally large values of f. In fact the f values for all of the radio atoms are high when compared with the rest of the plot, with the exception of radium F, radium-lead, thorium C′, lead from thorium, and uranium.

The electron content is relatively low in elements 39 to 51, and 59 to 63, the latter elements being praseodymium, neodymium, eka-neodymium, samarium, and europium, the former yttrium to antimony. From 34 to 42 the formula electron content is almost constant at about 12, as has been pointed out by Kossel. The slope of the plot from atomic number 29 to 84 is approximately 0.335 electrons per atomic number, or $^1/_3$, or on the average one *pair* of cementing electrons is added for each 3 α additions, while among the radioactive atoms this occurs on the average for 1.66 α changes.

Fig. 5 shows that whenever the atomic weight decreases with increasing atomic number, this is due to a sudden drop in the number of formula electrons, accompanied by a corresponding decrease in the cementing electrons in the nucleus.

An important deduction may be drawn from the cementing electron plot (Fig. 2) which gives the number of cementing electrons between atomic numbers 2 and 27 and between 82 and 92, as calculated from Equations 6 and 7, while from 28 to 81 only Equation 6 is used. *It will be noted that in the range between numbers 32 and 79 every even numbered element is represented by a peak in the curve, while every odd numbered element is represented by a trough,* though between atomic numbers 2 and 27 both the even and the odd numbered elements lie together on 2 horizontal lines, that is 2 lines which indicate a cementing electron content of 0 and 2, respectively. This suggests that while the atomic species represented between numbers 2 and 27 are largely pure, between numbers 32 and 79 either each even or each odd numbered element, or both, ceases to be a single pure species. An inspection of Equation 6 indicates that if an odd numbered element should happen to contain a considerable proportion of helium series atoms, Equation 6 would give too *low* a content of cementing electrons, as the equation is developed on the basis that odd

numbered elements consist entirely of members of the lithium series. If the *odd numbered element* should belong entirely to the helium series, then a result too low by 0.5 of a cementing electron would be obtained. On the other hand, the inclusion of either lithium or uranium series atoms in elements of even atomic number increases the result calculated from Equation 6 above the proper value (by 1.5 if the element contains only lithium series, or by 1.0 if it contains only uranium series atoms). It is of interest to note that in the range given the mean elevation of the even over the odd numbered points is 1.1 cementing electrons.

On the Existence of Alpha Particles in Complex Nuclei.

Two recent papers[1] have treated the structure of complex nuclei as if they are aggregates of positive and negative electrons which are not grouped into α-particles or μ or mass 3 particles, but are arranged in what seems to the writer of the paper, the most stable general arrangement for the number of positive and of negative electrons concerned. There are a number of facts and well grounded theories which seem to indicate that the α-particles largely preserve their identity in complex nuclei. Thus (1) α-particles are shot out from such nuclei, (2) the atomic weights and nuclear charges of pure species indicate that complex nuclei are largely built from groups with a weight of 4 and a charge of 2, and (3) while there is a change of weight and presumably of mass equal to 0.77% in the formation of one α-particle from 4 positive and 2 negative electrons, *the change of weight and mass when 3 or more α-particles unite to form a more complex nucleus, is so small that it has not been detected. Thus the α-complexes are extremely less stable than the α-particles itself.* It should be kept in mind, however, that there may be some loss of identity in the union into a complex nucleus, since there may be a partial redistribution of linkages with little change in the total energy, and, therefore, very little packing effect, that is loss or gain of mass.

The Search for Eka-Cesium and Eka-Iodine, and the Question as to the Existence of Elements of Higher Atomic Number than Uranium.

Serious attempts to separate eka-cesium have been made by Richards and Archibald,[2] by Baxter,[3] and by Dennis and Wyckoff.[4] So far as is known to the writer no extensive investigation has been carried out for the purpose of dicovering an element of higher atomic number than uranium. The positive charge on the nucleus of uranium has already a high value (92) and the resultant self-repulsion *may* be so high that if any atoms of higher atomic number have existed on earth at any time, they have already disintegrated so completely that they exist in such small

[1] Haas, *Physik. Z.*, 18, 400–2 (1917); Schmidt, *ibid.*, 20, 448–50 (1919).

[2] *Proc. Am. Acad.*, **38**, 443 (1903).

[3] THIS JOURNAL, **37**, 286 (1915).

[4] *Ibid.*, **42**, 985 (1920).

amounts that they cannot be detected by any ordinary means. That serious endeavors to discover such elements have not been made is probably due to a widespread, but unexpressed idea that such elements, if they exist, would be extremely radioactive, and so would have been detected in radioactive measurements on known materials. It seems worth while, in this connection, to consider certain facts in regard to the atoms of high atomic number.

Ordinary bismuth, which is probably a mixture of isotopes, though it may or may not consist largely of one atomic species, has a nuclear charge equal to 83. Above this there is no very stable species of atoms up to thorium, (90) the most stable isotope of which has an average life of 2.5×10^{10} years. It is also a fact that the most stable isotope of elements of even atomic number, has in general a greater stability than when the atomic number is odd. It would seem that if any such elements exist at all, they would be more likely to belong in the osmium, platinum, mercury, lead, or polonium groups, with possible atomic numbers 94, 96, 98, 100, and 102. Which of these would be the most stable is not indicated by the theory in its present state, though 98 or 100 would be indicated if the series above uranium should be similar to that below it.

With respect to eka-cesium and eka-iodine it should be noted that bismuth (83) has the highest atomic number which is odd with the exception of those atomic species which are descendents of the uranium or of thorium, and that both of the uranium series (radium and actinium series), and the thorium series as well, as now known, omit the atomic numbers 85 (eka-iodine) and 87 (eka-cesium), while both the uranium-actinium and the thorium series include every other atomic number from 82 to 92, inclusive. From this standpoint the discovery of either of these elements involves either the discovery of new branches of these radio series or of a new radio series, or else it involves the pushing upward of the odd numbered series from a nuclear charge of 83 (bismuth) to 85 and 87. The odd numbered descendents of uranium and thorium owe their odd nuclear charge to an odd number of negative nuclear electrons, which, it has been seen, leads to instability. That eka-cesium and eka-iodine are indicated by the Mendeleef periodic system does not show that they exist any more than it shows the existence of atoms with a nuclear charge higher than 92. On the other hand, eka-manganese, dwi-manganese, eka-neodymium, and eka-lutecium have atomic numbers which are small enough to give little reason to suspect their non-existence.

Mutual Electromagnetic Mass, the Relativity Effect, and the Question of a High Frequency Radiation Taken up or Given Off by Atom Nuclei.

According to one of the older theories the disintegration of radio atoms is due to energy absorbed in the form of radiation; in later years the energy

utilized has been supposed to be that stored up in the nucleus. The older theory has recently been revived by Perrin. If the atomic weights of the radio elements were known with sufficient accuracy, it should be possible to make a decision between these 2 theories, provided only one of these effects is operative. If the latter theory is considered, it is found that when interpreted in accord with the idea that energy has mass, it indicates that the change of mass, and presumably of atomic weight between uranium and lead, should be greater than the sum of the masses of the α-particles shot off, when the mass of the latter is taken when they are at rest. According to the relativity theory it should be 0.05 g. per gram atom greater in the specific case cited. According to the idea that the energy is absorbed from a penetrating radiation, the difference in the atomic weights need be no more, and from the point of view taken by Perrin, that the reaction is highly endothermal, would be less than the sum of the masses of the α-particles. The present atomic weights indicate a difference greater than that required by the energy storage theory, but this fact does not decide the question, since the accuracy of the atomic weights is at present too low for a definite decision. According to the radiation theory the different atoms of a pure atomic species would differ in mass slightly, according to their energy content, and would have a specially high mass just before they disintegrate. In later paragraphs the data will be discussed from the standpoint of the storage-relativity theory, though the writer is favorably inclined toward the idea that energy may enter and leave the nuclei by the action of some form of radiation.

The decrease in mass in the formation of one gram atom of helium nucleus is equivalent to 6.71×10^{11} calories according to the relativity theory. Earlier papers of the series show that the change of mass which occurs is 0.77%, and that when these α-particles combine with each other there is practically no change of mass. The question which now arises concerns the packing effect in other primary electron groups, such as the ν group (or lithium nucleus), and the μ group. If the atomic weights of lithium, determined by Richards and Willard, and of boron, determined by Smith, are considered as exact, then it is found that the packing effect in both would be abnormally high, amounting to a decrease of mass of 1.62% in the former case, and 1.69% in the latter if isotopes were absent which is improbable. If it is now considered that the lithium nucleus is the primary group of the odd numbered series, and that boron consists of one lithium plus one helium group, and that the packing effect in the latter has its normal value, that is that its weight is 4.00, then it is found that the lithium group in the boron nucleus has been subjected to the extremely large packing effect of 2.18%, while if it is assumed that the boron atom consists of two α-particles and

one ν group, the packing effect in the latter, calculated on a similar basis, amounts to 4.11%. The decrease of mass calculated for the apparent ν particle in the *lithium* nucleus is 2.75%. While packing effects of these magnitudes are not at all impossible, they do not seem very probable, when compared with those calculated in cases where the atomic weights are known with more certainty. Unfortunately, for this special problem, the nitrogen nucleus may or may not contain particles of mass 3, though it may be noted that it has the weight of 2 lithium nuclei, and the atomic weight of fluorine is not known with sufficient accuracy for our purpose though it is probably close to 19.00. The atomic weight of sodium is already so large that a small difference in the packing effect in the ν group (or lithium nucleus) contained in it, would not be noticed. However, the atomic weight of the ν group calculated from sodium comes close to 3.00. On the whole it seems probable that the packing effect in the ν group is of about the magnitude as in the α-particle. If the atomic weights 6.94 and 10.9 for these elements are sufficiently exact, which is almost certain it seems very much more reasonable to suppose that both lithium and boron contains small amounts of lighter isotopes, than to assume that such abnormally large packing effects as those cited above are real. According to the general theory presented in this paper, the value of N/P for the less abundant and lighter isotopes of lithium and boron should be 0.5, so the atomic weight of the lighter isotope of lithium should be 6, and that of the lighter isotope of boron should be 10.

If there were no packing effect in the μ group, its atomic weight would be 2.0156, while if this effect is equal to that in the α-particle, it would be 2.00. Since only one μ group occurs in any single atom, so far as can now be determined, and since the atomic weights have been determined accurately only for the heavier members of the μ or uranium series, no error of importance will be made by the use of the latter value.

TABLE III.—ATOMIC WEIGHT OF INTRA-ATOMICALLY COMBINED HELIUM.

In atom of even atomic No.	No. of α-particles.	At. wt.	Atom of odd No.	No. of α-particles.	At. wt.
Carbon	3	4.001	Sodium	5	4.000
Oxygen	4	4.0000	Aluminum	6	4.017
Sulfur	8	4.008	Phosphorus	7	4.003
(Argon)[a]	10	(3.99)	Vanadium	12	4.000
Calcium	10	4.007	Manganese	13	3.995
Titanium	12	4.008	Cobalt	14	3.998
Chromium	13	4.000			
(Iron)[a]	14	3.989			
Thorium	58	4.0026			
Uranium	59	4.0030			
Radium	56	3.9983			
Radio-lead (RaG)	51	4.0014			

[a] Probably argon contains an isotope of atomic weight 36, and iron of atomic weight 52.

As nearly as is now known, the atomic weight of helium itself is 4.000 (oxygen = 16). The weight of intra-atomically combined helium may be obtained by dividing the atomic weight of any pure atomic species by the number of α-particles contained in its nucleus. In cases where ν or μ groups are also present the weight 3.00 or 2.00 will first be subtracted. The preceding table contains values calculated in this way from the element weights (so-called atomic weights) which have been determined with considerable accuracy for elements which seem to consists mostly of a pure atomic species.

The average of the above results is 4.0013, for the 8 light atoms of even number it is 4.0008, for the 6 light atoms of even number 4.0022, and for the 4 heavy radio atoms 4.0013. Thus the average for the heavy atoms is very close to that for the light atoms.

Discussion of the Atomic Weights on the Basis of the Energy Storage Theory.

The mass equivalent of the heat given off in the disintegration of the radio atoms was calculated by R. Swinne[1] in 1913. Two years later an independent calculation of these values was made for me by W. D. Turner, and his values, which are practically the same as those of Swinne, will be used here. According to these calculations 8 α-changes in the uranium series give an energy change equivalent to 0.0519 g., or 0.0065 g. per α-change. In the thorium series 7 α-changes are equivalent to 0.0534 g. (0.0076 g. per α-change), and 5 disintegrations of the same type in the actinium series, to 0.0401 g. (0.0080 g. per α-change), all calculations being made for a gram molecule. Thus the mass effect per α-disintegration is greatest in the actinium, and least in the uranium series.

If the atomic weight of helium is taken as 4.000, and if it is assumed that all of the α-particles in the uranium nucleus give the same average energy of disintegration as in the radioactive series, then the loss of mass should be 4.000 plus 0.0065 or 4.0065 instead of the 4.0009 g. calculated from the atomic weights of uranium, radium, and radio-lead. In the thorium series the corresponding values are 4.0076 and 4.0023. Thus, on the average, there seems to be a smaller mass effect in the atoms as a whole than that calculated from the radioactive atoms alone.

The atomic weight assigned to thorium by the International Committee is 232.15. The weight of thorium lead should be this value minus 6×4.000 and also minus the relativity effect of approximately 0.05 g., or 208.10. That of radium-lead should be 238.175 minus 8 times 4 and also minus approximately 0.05, or 206.12. The lowest determination of the atomic weight of this lead obtained by Hönigschmidt is 206.06, and by Richards, 206.08, an average of 206.07, or a difference of 0.05

[1] Swinne, *Physik. Z.*, **14**, 145 (1913).

from the value calculated above. Hönigschmidt obtained 225.97 for the atomic weight of radium, and after making an allowance for the relativity effect, it is found that the determined difference in the atomic weights of uranium and radium is 0.19 units greater than the calculated deviation. It is, however, possible, as suggested by Piccard, that uranium contains an isotope of higher atomic weight.

It is quite evident from the above calculations that if the radioactive elements were to disintegrate completely into helium, the amount of heat given off per alpha disintegration could not, on the basis of the energy storage theory, continue to be as great as that which is liberated in the different radioactive series. This is just what would be expected, since the heat of disintegration becomes smaller as the period lengthens, that is as the atoms become more stable. It is of importance in this connection that the atomic weights of helium, and of the radioactive elements, should be determined with a considerably greater accuracy, since it is evident that the present atomic weights are not sufficiently accurate to indicate whether the disintegration of the radioactive elements is highly endothermal, as is claimed by Perrin in his radiation theory, or is highly exothermal, as indicated by the ordinary, or energy storage theory,[1] though the discussion given above shows that the most recent atomic weights of thorium and uranium indicate even a greater difference than that calculated by the storage theory, which, insofar as it has any value, is unfavorable to the theory of Perrin, though, on the other hand, the present atomic weights of radium and lead from radium are quite in accord with his theory.

The most important experiment which could be carried out in connection with the relation between the loss of mass and the relativity theory would be to weigh at intervals over a period of many years a carefully sealed and protected sample tube, containing an amount of mesothorium or radium equivalent in activity to possibly 10 g. of radium (or less with a delicate microbalance), so sealed that no α-particles and few β-particles could escape. The amount to be used could be calculated when the sensitivity of the balance and its accuracy have been determined. In this way energy could be allowed to escape in such amounts, that it should be

[1] Even on the basis of the theory of Perrin it is necessary to assume that the radioactive species of atoms are more unstable than the ordinary atoms, or else that their nuclei absorb more of the penetrating radiation. The relations between the periods of the radioactive elements have been shown in this paper to depend upon the relations between the number of positive and negative electrons in the nucleus, and upon other factors of a similar nature, which seem to indicate that the disintegration is related to the instability of the nucleus, rather than to the amount of energy picked up in the form of radiation. I would like to suggest that it is quite likely that a combination of the 2 theories may be found more in accord with the facts when they are sufficiently determined, than either Perrin's or the ordinary theory alone.

possible to determine its weight by difference. It is probable that this experiment has suggested itself to a number of workers, but the difficulty in the way of its execution is the great initial expense.

The Atomic Weights of Pure Atomic Species are Very Nearly Whole Numbers on the Basis of Oxygen as 16.

It is probable that the atomic weights of the light atoms which consist of only one species would be made more accurate by changing them to whole numbers. The atomic weights of nearly all of the light *elements*, with the exception of those where it is now almost certain that stable isotopes exist, are whole numbers, while from atomic number 28 to 80 they are no closer to whole numbers than they should be by the laws of chance, that is, this is the region of isotopes which are stable. The atomic weights now used for uranium and thorium are closer to a whole number than corresponds to chance, even though both of these elements contain isotopes. However, in this region the stability of one isotope is so much greater than that of the others as to make it so predominant in abundance that both of these elements occur as nearly pure species.

Nickel.

From the standpoint of atom-building nickel is one of the most interesting of all elements with the exception of hydrogen, helium, lithium, and the radioactive elements. Nickel, as is seen in Figs. 2, 3, 4 and 5, has an abnormally low negative-electron content, exceedingly low for its position in the system, and lies just at the beginning of the rapid increase in electron content. It is the first element in which the divisibility of the atomic weight by both 1 and 4 suddenly stops. It is the last of the abundant elements as the atomic number or weight increases, and it undoubtedly is a mixture of isotopes, as are most of the elements of higher atomic number. It should be possible to show the presence of the isotopes, most quickly by the positive ray method, and with much difficulty by diffusion. We are preparing to study the problem and especially the *isotopes of lithium* in this laboratory. Evidently no species of atoms in which the number of cementing electrons per atom is greater than 2 occurs abundantly either in the meteorites or on earth.

Summary.

1. The negative electrons in the nuclei of atoms are largely associated in *pairs*, either as *binding* or *cementing* electrons. The pairs of binding electrons serve to bind together a number of positive electrons into a primary group or particle. The *most abundant by far of all of these is the α-particle*, which consists of 2 negative and 4 positive electrons, and, therefore, has a net positive charge of 2. Its formula is $(\eta_4{}^+\beta_2{}^-)^{++}$, where η^+ is the positive and β^- the negative electron. This α-particle probably makes up about 90 or more % of all known material in the meteorites. A second less

abundant group is the $(\eta_2{}^+\beta_2{}^-)^\circ$ or μ group, which has a zero net charge, and probably makes up more than half of the rest of known material. It is, of course, possible that this may occur only in union with the constituents of an α-particle as $(\eta_6{}^+\beta_4{}^-)^{++}$, or meta-helium, but the former hypothesis has the advantage of greater simplicity, with no loss of usefulness. Thus nearly all of the material which consists of complex nuclei, exists in the form of groups made up of pairs of negative binding electrons, *together with an even number of positive electrons.* Isotopes of higher, differ from those of lower atomic weight by the presence of a single μ group, or of one or two helio groups, or of both a helio and μ group. The term helio group is used to designate an α-particle together with two negative electrons, which may be said to "cement" it to the nucleus of the atom. The above discussion of the abundance relations of the primary electron groups is based on data obtained from the composition of the meteorites and of the earth. Some of the stars and nebulas probably contain large amounts of hydrogen, and in such a case the relations of abundance given could be expected to hold only for the complex nuclei, and not for the positive electron (hydrogen nucleus). Atoms of odd net nuclear charge are relatively rare. They, also, consist mostly of α-particles, but the odd value of the net charge is caused by the presence of one odd numbered ν group $(\eta_3{}^+\beta_2{}^-)^+$ in the nucleus of each atom of odd atomic number. Of the light atoms only beryllium and nitrogen seem to contain an odd number of negative nuclear electrons, and these elements are not abundant. Even among the radioactive nuclei there are only a few which contain an odd number of negative electrons, and they are very unstable.

2. The α-particle may be assumed to be electrically negative in most of its exterior, but to have a net positive charge of 2. Such particles would repel each other at all ordinary and small distances, and would only attract when brought extremely close together in such a way that electrical couples are able to produce a greater attraction than the repulsion due to the net charge. Neither a single positive or a single negative electron can attach itself to such a particle, nor does it seem that 2 such particles will unite, but from 3 to 8, and also 10, but not more than 10 α-particles unite to form a complex nucleus in which the mass is twice the charge, and the ratio of negative to positive electrons is 1 to 2, or 0.5. *This ratio is of great importance in relation to the stability of nuclei.* Eleven positively charged α-particles will not alone unite[1] to form a complex nucleus, since their positive charge, 22, is so large that the attractive action of the α-particles is not able to overcome the repulsive effect of this posi-

[1] It is likely that this is not strictly true, since it is probable that as many as 14 α-particles unite to form the nucleus of the lightest nickel atom without the inclusion of any negative α-cementing electrons. However, the above statement is true if only the most abundant isotope of each element is considered.

tive charge. *However, more positive α-particles will add on if at the same time the ratio of negative to positive electrons is increased. The ratio is increased by the addition of an α-particle together with 2 cementing electrons,* or what has been called a helio group. This helio group increases the ratio of negative to positive electrons by a sudden jump from 0.5 to 0.55 (approximately). The increased ratio makes it possible for an extra positive α-particle to attach itself, so two α-particles add on in this one step, and in addition 2 cementing electrons. *Thus the function of cementing electrons is to keep the nucleus stable as its self-repulsion, due to its increasing positive charge, increases, by also increasing the relative negative electron content. This causes 2 α-particles to add on, so the atomic weight increases by 8, instead of the normal 4.* More α-particles then add on, decreasing slightly the N/P ratio, until the repulsion of the positive charges again becomes so high that a second increase in the ratio becomes necessary, when a second pair of negative cementing electrons adds on, and the atomic weight again rises with abnormal rapidity. Thus if the atomic number is plotted on the X-axis, and the number of cementing electrons on the Y-axis, the plot consists of a series of horizontal lines, each of which has at its right a vertical rise of 2-cementing electrons, followed by another horizontal line; that is, a series of steps with equal vertical spacing. The N/P ratio plot is somewhat similar, but the steps all slant downward slightly toward the right.

3. The writer shows that 4 series of atoms are now known; the *helium-thorium* series, the *meta-neon-uranium* series, the *lithium-cobalt* series, and the *meta-chlorine* series. The relation of these 4 series to his periodic system is briefly discussed.

4. *The atomic weights of all pure species of atoms are very close to whole numbers when calculated on the basis of oxygen as 16.* This is practically the statement made by Harkins and Wilson in 1915. At that time it was plainly seen that this rule holds for all light atoms, and for the heavy atoms far up in the series, but there was some doubt as to the magnitude of the relativity effect, that is the change of mass due to packing. The atomic weight determinations of Richards and Hönigschmidt show that these atomic weights are either whole numbers, or else very slightly above whole numbers. If the energy given off on disintegration comes from potential energy which remains stored in the nucleus as long as it exists, then it is probable that there is a slight drift away from whole numbers in some parts of the series on account of the mass associated with the energy changes involved, which amounts to about 0.05 g. per gram atom in each radio series, involving 6 or 8 α-changes. If the energy comes from without, as is assumed in the recent theory of Perrin, the loss of mass of the complex atom might be equal to or more probably less than the sum of

the masses of the α-particles ejected, the masses being taken when the α-particles are at rest.

5. Tables are given for the total positive, total negative, α-cementing, excess, and formula, electron content of the nuclei of all known atoms, and the more important relations which are apparent, are pointed out.

6. When the net positive charge on the nucleus of an atom rises higher than 28 it seems probable that the resultant self-repulsion due to the unneutralized positive charge becomes so great that relatively few atoms of this class can be formed. Such atoms are relatively rare and probably do not make up more than one-thousandth of the total material of the earth and the meteorites, though they constitute more than $^2/_3$ of all of the elements, and probably more than $^3/_4$ of the atomic species. In all abundant atoms the ratio $\frac{\text{Total number of negative electrons}}{\text{Total number of positive electrons}}$ is 0.5 or only very slightly above that value. Thus it is 0.5 in oxygen, 0.5 in Si_{28}, 0.536 in Fe_{56}, 0.5 in Mg_{24}, 0.5 in sulfur, 0.5 in Ca_{40}, and 0.522 in $Ni_{m58.68}$, the 7 most abundant atomic species. Here m58.68 indicates that all of the isotopes of nickel, with a mean atomic weight of 58.68, are included. The atoms become rare as soon as the cementing electron content of the nucleus rises above 2, that is it seems probable that no number of cementing electrons is able to impart stability of a high order to a nucleus whose positive charge is greater than 28.

7. In general nuclei which contain an odd number of positive electrons are rare, and those which contain an odd number of negative electrons are much rarer still. This means that atoms of odd nuclear charge are rare, and when they exist the odd number of the charge is almost always due to an odd number of positive, not of negative, electrons.

8. The peculiar relations of nickel are pointed out. It is undoubtedly a mixture of isotopes. Nickel has an extremely low mean negative electron content in its nucleus, and, therefore, has a low atomic weight for its position in the system.

9. Nitrogen, and also scandium if its most recent atomic weight determination is correct, are not members of any of the 4 ordinary series of atoms listed above.

10. Nickel, potassium, calcium and iodine, have an abnormally low nuclear content of negative electrons, while the number of such electrons in argon, tellurium, barium, and to some extent in selenium, is relatively very high. It will be noted that most of these abnormally high and low values lie in close proximity to the 3 reversals in the atomic weight increase with increasing atomic number.

11. While there are only 92 elements, in the limited sense in which the term is now used, in the ordinary system, there are probably 300 or more different species of atoms, which, in a broader sense of the term, are truly

elements. It is obvious that the word element is now used in a very arbitrary sense to denote all atoms in whose nuclei the difference $P - N$ (where P stands for the number of positive, and N, the number of negative electrons) has the same value.

12. The plot of the cementing electron content of atom nuclei, indicates that nearly all of the elements from atomic number 32 (Ge) up to 79 (Au), are mixtures of isotopes, and that in general there are probably several isotopes present in considerable percentages in each element. Other facts indicate strongly that the region in which isotopes become extremely prominent begins with atomic number 28 (nickel).

13. The atomic weights of the light elements are mostly whole numbers on the oxygen basis, and in the case of the elements of even number are divisible by 4. This indicates, according to the theory of the earlier papers of this series, that isotopes do not occur in *high percentages* in any of these elements, with the exception of neon, chlorine, magnesium and silicon (possibly in aluminum). From atomic numbers 28 to 80 the atomic numbers are no closer to whole numbers, and the atomic weights of the even numbered elements are no more nearly divisible by 4 than corresponds to the laws of chance. Among the radioactive elements, however, both the approximation to whole numbers, and divisibility by 4 in the case of the thorium series, again become prominent.[1] In other words, the general relations found earlier by the writer hold both for the light and for the very heavy atoms, but not for those in the intermediate range. This is easy to understand from the viewpoint taken in the earlier papers. The light atoms are made up from relatively few α- and other particles, and not many stable arrangements could be expected under these conditions. As the number of particles in the nucleus increases, the number of possible arrangements increases, and in the region of stable atoms the number of stable arrangements increases and becomes large between atomic numbers 28 and 84. However, in the region of the very heavy atoms, while there are many possible arrangements, even the most stable of these is already unstable, so it could not be expected that the less stable atoms would have such long period of existence as to give them prominence by their abundance. Thus, among the radioactive atoms, in spite of the presence of numerous isotopes, there is seldom more than one species of atoms in an element which has sufficient stability to be largely represented in the atomic weight.

14. The atomic weights of lithium and boron indicate the existence of lighter isotopes, and the theory indicates that in them the ratio N/P should be 0.5, so the atomic weight of the lighter lithium (present probably to the extent of about 6% in lithium) should be 6, while that of boron should be 10. Also an isotope of iron, presumably of atomic weight 52;

[1] A table illustrating these facts is presented in the *Phys. Rev.*, **15**, 81 (1920).

a higher isotope of calcium, possibly atomic weight 44; and a higher isotope of potassium, are all indicated, in addition to several isotopes of nickel as mentioned above, *presumably* with atomic weights 56, 60, and *possibly* other values including 58.

15. Attention is called to the suggestion made in an earlier paper in the *Physical Review* that atoms of zero atomic number may exist and be of importance in atom building. Such atoms might have masses 4, 3, 2, and 1, and possibly other values, and they would contain no non-nuclear electrons, so they would have no chemical, and almost none of the ordinary physical properties, aside from mass.

16. The general theory of nuclear structure presented in the earlier papers of this series indicates, as has been pointed out by N. F. Hall,[1] that *in general the number of isotopes present in elements of even atomic number should be considerably greater than in elements of odd atomic number.* According to the theory presented in the present paper it is probable that this relation may not hold true for atoms of low atomic number, but it may be expected to be valid beyond atomic number 26 and possibly beginning with still lower atomic numbers. It is of interest to note in this connection that, *since this paper was submitted* Aston has found 6 isotopes of krypton and 5 of xenon, elements of even number, but only 2 for bromine (79 and 81) and only one atomic species for arsenic, both elements of odd atomic number.

17. It is of interest to note that the formula for the atomic weights presented 5 years ago does not give the atomic weights of all of the isotopes of an element, but it picks out that of the most abundant isotope. It does this in the range of the light atoms (atomic numbers up to 27) at least. Thus for neon, with atomic weights 20, 22, and probably 21, it designates the species of atomic weight 20, which makes up about 90% of the element neon. The formula is

$$W = 2(n + c) + {}^1/_2 + {}^1/_2(-1)^{n-1}.$$

18. The paper states that the *element chlorine has been separated into isotopes of separate atomic species* by Mr. C. E. Broeker and the writer. We suggested in *Nature* that in addition to Cl_{35} and Cl_{37}, there seemed to be some indication of another chlorine of higher atomic weight. The atomic weight of this last chlorine was later found by Aston to be 39, but the percentage in ordinary chlorine is very small. In the experimental investigation Dr. W. D. Turner did most of the early, and Mr. T. H. Liggett some of the later work. The separate publication of this work has been delayed by the death of Mr. Broeker.

19. The whole number relation of the atomic weights when oxygen as 16 is taken as a basis, may be stated in the form that in all known complex nuclei the positive electron has the weight 1.000 ± 0.001, while the free positive

[1] *Loc. cit.*

electron has the mass 1.007, very nearly. This might be stated more accurately by giving the weight of the average electron pair, positive plus negative electron, as 1.000 ± 0.001 in any complex atom, but as 1.0077 in hydrogen itself where the positive electron is *free* and not bound. The constancy in the packing effect may be a characteristic of the positive and negative electrons themselves.

20. In considering the foregoing paper it should be realized that it develops some *very general relations*, such as those involving the ratio of negative to positive electrons in the nucleus (N/P), which are very likely to prove entirely valid, and that it gives very *special details*. such as formulas for nuclei, etc., which are not expected to fit the facts in every special case, but are only intended to illustrate the general relations and to give a specific theory to which the facts as they are discovered may be related. The nucleus is so complex that it is possible that nuclei of atoms of the same element may have a different composition with reference to the particles of masses 4, 3, 2, and 1, even when their atomic weights are the same. It is quite likely that the relations concerning the ratio N/P are much more general than those relating to the specific formulas. Thus the important feature about the oxygen nucleus *may* be that in it the ratio N/P is 0.5, and not so much that it consists of 4 α- particles.

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THE X-RAY FLUORESCENCE OF CERTAIN ORGANIC COMPOUNDS.

BY H. S. NEWCOMER.

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In a previous[1] paper I pointed out that it might in the future become possible to make use, biologically, of a localized transformation of penetrative radiant energy with very feeble abiotic properties (X-ray), into a much more active, although less penetrative type of ray (ultra-violet) through fluorescent substances as intermediaries. It was shown, in fact, that fluorite under the influence of the X-ray emitted bactericidal rays. Progress from this point demanding that we have for use in place of the insoluble fluorite a similarly active soluble substance, a survey was made of a large number of substances, for the most part organic chemicals, to see which of them might fluoresce under the influence of the X-ray.

Kunz and Baskerville[2] have examined the action of radiation on 13,000 minerals in the collection of the American Museum of Natural History. Their most noteworthy observations were the variability in like minerals

[1] H. S. Newcomer, *J. Exp. Med.*, **26**, 675 (1917).

[2] G. F. Kunz and Chas. Baskerville, *Science*, **18**, 769 (1903).