Nuclear Binding Energies

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THE numerous atomic species or nuclides, which differ widely in their various properties, can be considered, whether they are formed in nature or whether they are produced artificially, as nuclear chemical compounds made of a few nuclear building stones, the so-called "nucleons." The first indication that this is so, as well as the first information concerning an important property of the nucleons—namely, their atomic weight—was given by the well-known law of integers of Aston. This law says that the weights of nuclides are practically integers, if they are expressed in atomic weight units. Naturally, the same also holds true for the sum of different nuclidic weights or, in other words, for the molecular weights of pure chemical compounds.

The fact that nuclidic weights are integers is the most striking feature in photographs of mass spectra; it is also the thing that makes these spectra so different from the patterns of optical spectra. Figure 1 shows sections of a photographic plate with a sequence of four mass spectra obtained in our mass spectrograph. The exposures were taken consecutively under otherwise constant conditions; only the time T of exposure was increased by a factor 3 each time, from 2 min 15 sec to 60 min 45 sec. A canal ray tube, into which a mixture of illuminating gas and air with a small amount of helium was introduced, served as an ion source, so that lines are present at almost every integer from the mass number $A=4$ to the end of the plate at $A=41$.

In the mass spectra with long exposure, one can also recognize lines between two integers, for example at $A=13\frac{1}{2}$ or at $A=13\frac{1}{3}$. But these also prove the law of integers, because they correspond to multiply charged ions, namely $^{27}Al^{++}$ (originating from the cathode of the discharge tube) and $^{12}C_2H_3^{++}$ or $^{40}A^{+++}$, respectively; since in the mass spectrograph one really determines the charge-to-mass ratio of the ions, multiply charged ions result in lines that correspond to one-half or one-third of their weight. The lines at $A=6$, 7, and 8 also correspond to such doubly charged ions—namely, to those of the nuclides ^{12}C, ^{14}N, and ^{16}O, which are here the most abundant ones. Especially characteristic in the exposures of the sections of the plate reproduced in Fig. 1 are the two groups that start at $A=12$ and at $A=24$, respectively. The first contains, next to each other and partly superimposed, the isotopes of the elements C, N, and O and their hydrides—that is, the dissociation products of CH_4, NH_3, and OH_2. The second group is formed mainly by C_2 and its hydrides, that is, by the dissociation products of C_2H_6; at $A=28$, the abundant molecular ions of CO and N_2 occur in addition, which makes this line appear particularly intense.

From the fact that their weights are integers, it was immediately concluded that the nuclides are built up from nucleons with an approximate atomic weight $A=1$. Indeed, one would need only one type of nucleon if it were not well known that the nuclei of the neutral nuclides have electrons bound in their shells, again in integral numbers, corresponding to the atomic numbers in the periodic system. One must therefore conclude that the nucleon of weight $A=1$ occurs in two manifestations or quantum states, once with a single positive charge as a proton and once uncharged as a neutron. The three nuclides $^{40}_{18}A$, $^{40}_{19}K$, and $^{40}_{20}Ca$, for example, which all happen to occur in nature, contain 40 nucleons each; but of these (according to the atomic number of these three elements indicated by the lower index) 18, 19, and 20, respectively, are protons, whereas the remainder, namely 22, 21, and 20, respectively, are assimilated as neutrons by the nucleus in question.

But the most interesting fact for our subject (*1*) is that Aston's law of integers can claim validity only as a more or less rough approximation. This, by the way, applies also to Lavoisier's law of chemistry. Even in ordinary chemistry the molecular weight of each exothermic compound must always be smaller than the sum of the atomic weights of the elements of which it is composed. In the process of compound formation, energy is released in the form of heat, and—according to Einstein's fundamental law of the equivalence of mass and energy—to each amount of energy there is a corresponding amount of mass, and vice versa. It has not yet been possible, of course, to measure the loss of mass ΔM, which corresponds to the binding energy (B.E.) in chemical reactions because, owing to the large conversion factor, this mass defect is extremely small. We have $\text{B.E.}=\Delta M \cdot c^2$, where c represents the velocity of light. The experimental situation is quite different, however, in the case of those binding energies that link the nucleons in nuclides. These nucleochemical binding energies surpass the ones in ordinary chemistry by a factor of about 1 million, and the corresponding loss of mass is therefore measurable in mass spectrographs of high resolving power.

The fact that Aston was able at all to state the law of integers proves something very strange. Indeed, each nucleon in its bound state must have an atomic weight of about 1.000—and this holds very nearly true for all nuclides heavier than about ^{16}O—but the atomic weights of the proton (neutralized as a hydrogen atom) and of the neutron amount to

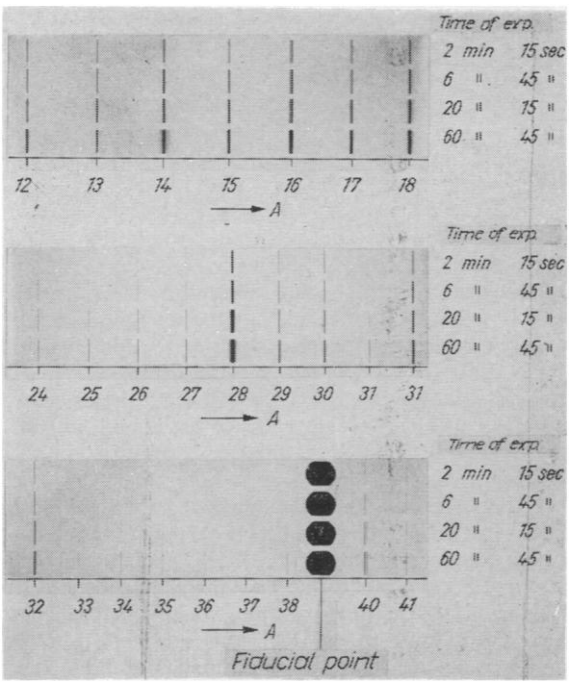

Fig. 1. Aston's law of integral mass numbers A. Sections of a photographic plate with mass spectra taken consecutively with increasing time T of exposure under otherwise unchanged conditions.

1.008 and 1.009, respectively. Each nucleon has therefore suffered the same average loss of mass of about 8.5 millimass units (mmu) in its bound state, or, expressed differently, the binding energy per nucleon has the same value, about 8 Mev in almost all nuclides, if we convert into the units of energy customary in nuclear chemistry (1 mmu = 0.931 Mev). If the mass defect per nucleon were not nearly constant for all nucleons above ^{16}O, or if it had a value different from the one just mentioned, then the nuclidic weights and the integers would get out of step for nuclides containing approximately 100 nucleons.

The binding of the nucleons in a large atomic nucleus is therefore quite similar to the binding of molecules in a condensed phase, such as a crystal or a liquid; that is, the total binding energy is, in first approximation, proportional to the number of molecules or of nucleons, respectively. This fact is the basis for the liquid-drop model of the nucleus. It is considerably substantiated by the result derived from other observations that the volume of the atomic nucleus is proportional to the number A of nucleons, or, to say it differently, the average density of the atomic nucleus remains constant for all nuclides throughout the whole periodic system. In addition, this result suggests that, in contrast to the situation existing in the atomic model, the material that forms atomic nuclei must be considered as rather similar to an ordinary liquid, in which the individual particles (nucleons) are packed closely together and maintain a constant density, independent of the size of the sample. One may therefore speak of a universal nuclear liquid that forms little droplets of various sizes and usually of spherical shape.

In 1927 Aston published his measurements of the deviations of the nuclidic weights from integers in the form of a packing fraction curve that reflects the variation of the binding energy per nucleon with the number A of nucleons in the various nuclides. In addition, the deviations from the constant value can be understood easily by means of the liquid-drop model. In heavy and large nuclei, there are, apparently, many more nucleons surrounded by others than there are in the light nuclei, where comparatively more nucleons are on the surface. In the light nuclei the (positive) surface tension per nucleon counteracts therefore the (negative) energy of cohesion. With a larger number of nucleons, the value of the binding energy per nucleon would be larger; the value for an infinitely large volume is reached asymptotically. In the case of heavy nuclei a different effect again has a loosening influence on the cohesion—namely, the Coulomb energy of the protons, which steadily increase in number and repel each other more and more. The binding energy per nucleon therefore reaches a maximum value in the neighborhood of $A = 60$. Considering energy alone, practically all heavy nuclei would be unstable against fission and all light nuclei would be unstable against fusion, but the reaction rates for these processes are so low that they are effectively zero. Fission produced by neutron capture by uranium and thermally induced fusion of hydrogen in the interior of stars (or on Bikini) can be considered exceptions.

The fusion of 4 nucleons to ^4He and the binding of ^4He onto ^{12}C, which results in the formation of the nuclide ^{16}O, liberates such a large amount of energy that the corresponding Einsteinian mass defect was already made visible in 1927 by Aston in the form of two neighboring but separated lines (doublet) for the molecular ion $^{12}CH_4$ and the atomic ion ^{16}O, both of mass number 16. Such a mass spectrum is reproduced in the bottom part of Fig. 2. After we, together with R. Herzog, had shown in 1934 how to calculate and build mass spectrographs according to ion-optical considerations, we were soon able (in 1938) to make photographs like the one shown in the middle of Fig. 2. It shows, in a subsequent enlargement, a picture of the same doublet that has now become a triplet by addition of the line $^{14}NH_2$, which divides the distance between the lines $^{12}CH_4$ and ^{16}O unequally. How far one can go with such enlargements and how accurately one can measure the distances between such multiplet lines and thus the mass equivalents of differences in the binding energy is finally limited only by the resolving power that can be attained. In our apparatus (*2*)—which in the last 20 years has undergone numerous improvements of a constructional and technical nature without any changes in the principle or even in the dimensions of the first arrangement—the resolving power now amounts to 100,000, that is two masses which differ by

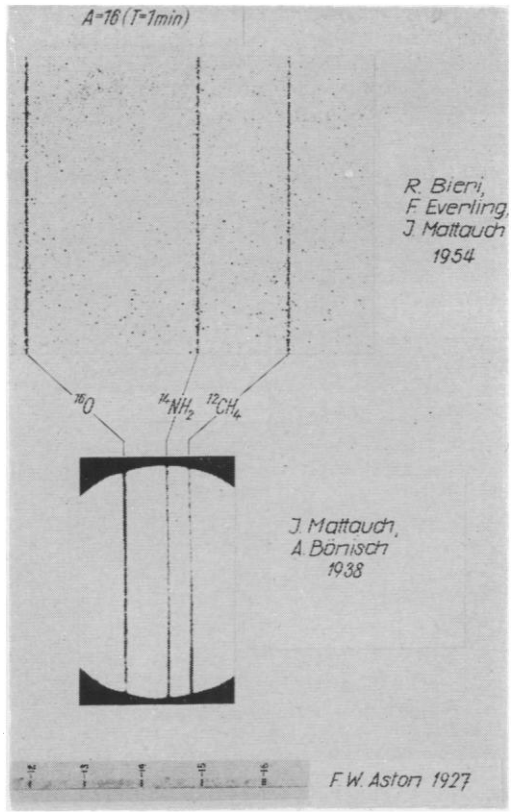

Fig. 2. Mass-spectrographic doublet ($^{12}C^{1}H_4 - {}^{16}O$) at $A = 16$. Showing the mass defect of the fusion of 4 nucleons to ^{4}He and the binding of ^{4}He onto ^{12}C leading to the formation of ^{16}O. Historically this is the first case of two well-resolved lines at the same mass number.

only 0.001 percent still appear as two separate lines. This corresponds in the middle of the plate to a line width of about 1 μ. The top part in Fig. 2 demonstrates the present state of the experimental technique. By means of such magnifications it is, of course, not possible to resolve unseparated lines or to add lines by magic; on the contrary, weak lines often disappear in the process.

Figure 3 shows in its lower part a multiplet at mass number 20, which we succeeded in obtaining in 1938 and which—today we might say unfortunately—was taken over by the authors of various textbooks. We find consolation in the thought that these books—for reasons that lie with the authors—are so popular that it will soon be possible to print the upper part of Fig. 3 in new editions. The advancement in resolving power can be seen not only in the larger number of resolved lines (10 instead of 6) but especially in the separation of the various heavy water molecules, for example. In the lower picture the line designated $^{18}OH_2$ appears as a weak, only poorly resolved satellite of the very intense line for $^{16}OD_2$. Today (upper picture of Fig. 3) the two are widely separated from each other, and in between them the very weak line for ^{17}ODH becomes visible. In 1938 we would not even have been able to exclude with certainty the possibility that the line designated as $^{18}OH_2$ was not produced partly by ^{18}OD also. Today the lines corresponding to these two molecular ions also appear separated by many times the line width. Naturally, one cannot hope to obtain in a case of 10 lines at the same mass number (we have sometimes obtained even more) all lines of the multiplet in equal intensity, especially if isotopic nuclides that are not at our disposal in 100-percent pure form are involved. For the remaining photographs I should like therefore to ask indulgence.

The simple liquid-drop model represents, surprisingly, many properties of the atomic nuclei very well. As is true of any model, its use should not, however, be stretched too far. As in the structure of the electron shell, which is so different in other respects, there appear certain closed shells in the process of building up the nuclei from nucleons. In the form of double and quadruple periodicities such filled shells were long known and even expected on the basis of the Pauli principle. Since we know that two kinds of nucleons exist, one charged and one uncharged, each of which can be built in with a spin of $+\frac{1}{2}$ or $-\frac{1}{2}$, there is always room for exactly 2, or even better, 4 nucleons (2 protons and 2 neutrons, each type with both spin orientations) in one shell. This is the reason why the formation of the ^{4}He-nucleus, which repre-

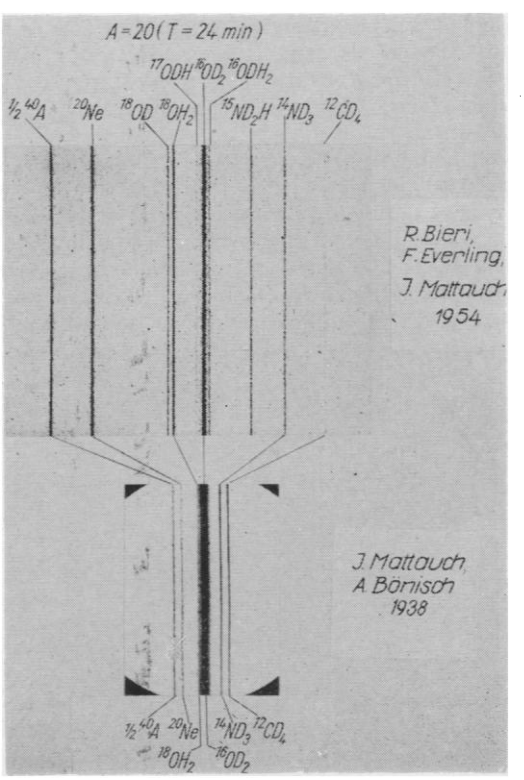

Fig. 3. Mass-spectrographic multiplet at $A = 20$ showing the increase in resolving power reached during the past 16 years.

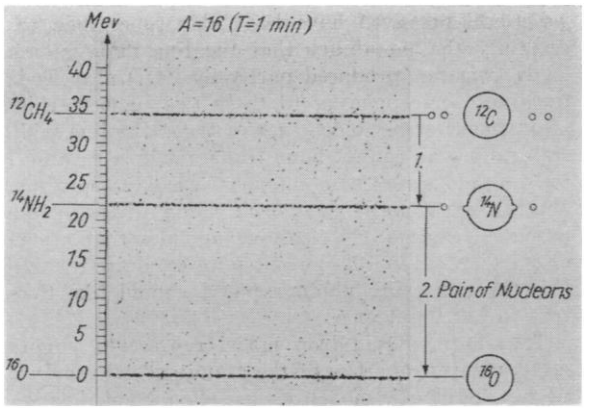

Fig. 4. Binding energies of successive pairs of nucleons leading from ^{12}C over ^{14}N to ^{16}O.

sents the first closed shell with 4 nucleons, yields the abnormally high energy of 28 Mev, an energy so high that to date we defray all our energy consumption on earth from it with the sole exception of the energy gained by nuclear fission.

In the last decade another type of closed shell has been reported in the papers of the Heidelberg group (Haxel, Jensen, and Suess) and also simultaneously by American scientists (Maria Goeppert-Mayer and others). These closed shells occur at certain proton and neutron numbers, which, at first, because an explanation was lacking, were designated as magic numbers. Filled shells are characterized by the appearance of a certain saturation of the enormously large binding forces that exist between the nucleons at small distances. Such nuclei show a definite disinclination to bind a new nucleon. The last nucleon that terminates the closed shell is bound, however, with an energy noticeably higher than the average. In an energy-level diagram the levels corresponding to the closed shells are therefore displaced slightly downward in comparison with the others. This can be illustrated with some of our multiplets, which if rotated

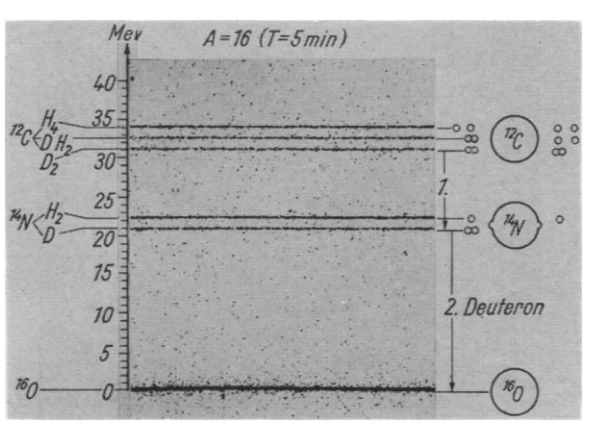

Fig. 5. Energy release by the formation of the deuteron and the binding energies of successive deuterons leading from ^{12}C over ^{14}N to ^{16}O.

by 90° represent nuclear energy-level diagrams, where instead of using the drawing pen, the mass spectrograph is used directly for drawing in the levels.

Figure 4 shows the triplet of Fig. 2 rotated 90° furnished with an energy scale and interpreted as an energy-level diagram. Special attention should be given to the nuclei indicated as large circles in the center of the rough model pictures on the right side, and one should interpret the chemically bound hydrogen atoms only as a far-distant "store" of nucleons from which we draw in order to build up the central nuclei further. In order to be able to interpret the photographs at all as energy-level diagrams, it is necessary to have enough nucleons in the "store" so that one is able to complete the nucleons of the central nuclei to the number A indicated on top of the photographs. We start with a multiple of ^4He—namely the ^{12}C nucleus—and we see that the building in of the first pair of nucleons liberates considerably less energy than the one of the second pair, which leads to the ^{16}O nucleus and therefore to a closed shell. In order to be accurate, we have to increase both mass differences ($^{12}CH_2 - {}^{14}N$) and ($^{14}NH_2 - {}^{16}O$) by the difference ($^1n - {}^1H$) = 0.844 mmu = 0.786 Mev in order to obtain the correct mass equivalent of both energy amounts—12.5 and 22.9 Mev, respectively—because both times a proton has been "assimilated" as a neutron by the central nucleus, since obviously we can have only the chemically bound hydrogen atoms in the "store." The difference ($^1n - {}^1H$) corresponds, however, in our photographs to approximately four to five line widths only and can be disregarded in a qualitative approach.

For the recording of the next energy-level diagram (Fig. 5) by the mass spectrograph we added deuterium gas to the discharge tube, so that all combinations might occur in the hydrides that can result through substitution of 1 D atom for 2 H atoms. The equidistant intervals ($^{12}CH_4 - {}^{12}CDH_2$), ($^{12}CDH_2 - {}^{12}CD_2$), and ($^{14}NH_2 - {}^{14}ND$), increased, however, by the difference ($^1n - {}^1H$), yield in each case the mass equivalent (quite small, by the way) of the binding energy of the deuteron (2.2 Mev). Here it can be seen clearly how, according to Einstein's law, the mass is reduced each time by the same amount, if fusion of 2 nucleons into 1 deuteron occurs in the "store." Furthermore, it is possible to read directly (arrows in the energy-level diagram) the energy amounts (10.3 and 20.7 Mev) that are released by the successive binding of preformed deuterons. The first energy amount is only half as large as the second.

The photographs for the remaining figures were achieved only with the help of samples that contained enriched amounts of the rare isotopes ^{13}C, ^{15}N, and ^{18}O (*3*). In Fig. 6 we are able to follow easily the binding of successive single neucleons to the central nucleus as well as the numerous fusions of nucleon pairs to deuterons in the "store." The mass equivalents of the corresponding binding energies are again marked in the diagram by arrows, and

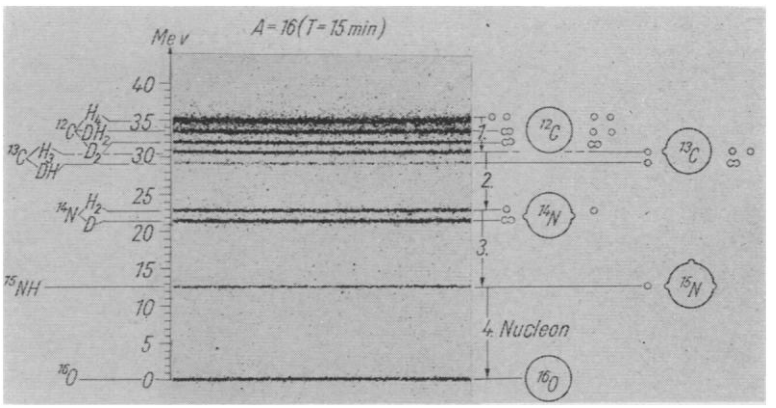

Fig. 6. Binding energies of successive nucleons in the sequence ^{12}C, ^{13}C, ^{14}N, ^{15}N, ^{16}O.

it can easily be seen that the first and the third each have to be increased by the difference $(^1n - ^1H)$ in order to obtain the correct result, for the first and the third arrows obviously combine isotopic nuclides. After the building in of each second nucleon a certain closed shell becomes apparent, for the even nucleons release considerably more energy than the preceding odd ones. The corresponding amounts are in the sequence of Fig. 6: 4.9_5 and 7.5_5 Mev as well as 10.8 and 12.1 Mev.

In Fig. 7 we are able to follow the same occurrence two mass units higher. The nucleon "stores" are everywhere increased by 2, if this is compatible with the chemical compounds. Again the picture is threaded through like a railroad yard by the standard gage parallel tracks so typical for the formation of deuterons, which can occur here in the "stores" of the nuclei ^{15}N and ^{16}O also. From the nucelus ^{12}C to the nucleus ^{16}O nothing was changed in the binding energies of successive deuterons and nucleons. Their amounts are identical with those that we obtained from the previous pictures. However, the closed shell at ^{16}O becomes very distinctive. This nucleus particularly is saturated and only little disposed to bind further nucleons. The next pair of nucleons, which leads to ^{18}O, consists of 2 neutrons built in with opposite spin. Its binding energy is given by the interval $(^{16}OH_2 - ^{18}O)$, which in this case has to be increased by twice the difference $(^1n - ^1H)$, in order to obtain the correct value of 12.2 Mev. It is somewhat smaller than the previous value for the binding energy of the nucleon pair taken from Fig. 4, which led from ^{12}C to ^{14}N.

The particular disinclination of the ^{16}O nucleus to bind even the very next nucleon is shown even more clearly in Fig. 8. The multiplet shown in Fig. 3 is prepared here as an energy-level diagram, and the pattern allows one to recognize in nearly all its details the building up of two successive shells of four. First of all we observe again the two deuteron steps leading from ^{12}C to ^{16}O, as shown in previous pictures. The interval between the two weak lines for $^{16}ODH_2$ (hydrides, not very familiar to the chemist but known for a long time to mass spectroscopists) and ^{17}ODH increased by the $(^1n - ^1H)$ difference gives the binding energy of the first neutron to the ^{16}O nucleus which represents the fifth nucleon, counting from ^{12}C. Its value—4.1 Mev—is smaller by 0.8 Mev than the corresponding value of the binding energy of the first neutron in the ^{12}C nucleus. The binding energy of the second neutron that leads to ^{18}O—that is, the step indicated in Fig. 8 as the sixth nucleon increased by the $(^1n - ^1H)$ difference—amounts to 8.1 Mev. It is approximately 0.5 Mev higher than

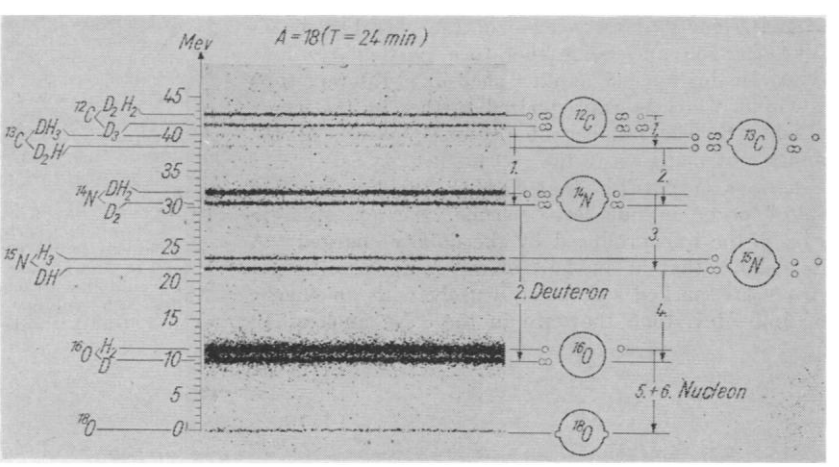

Fig. 7. Binding energies of successive deuterons and nucleons in the sequence that leads from ^{12}C over ^{16}O to ^{18}O.

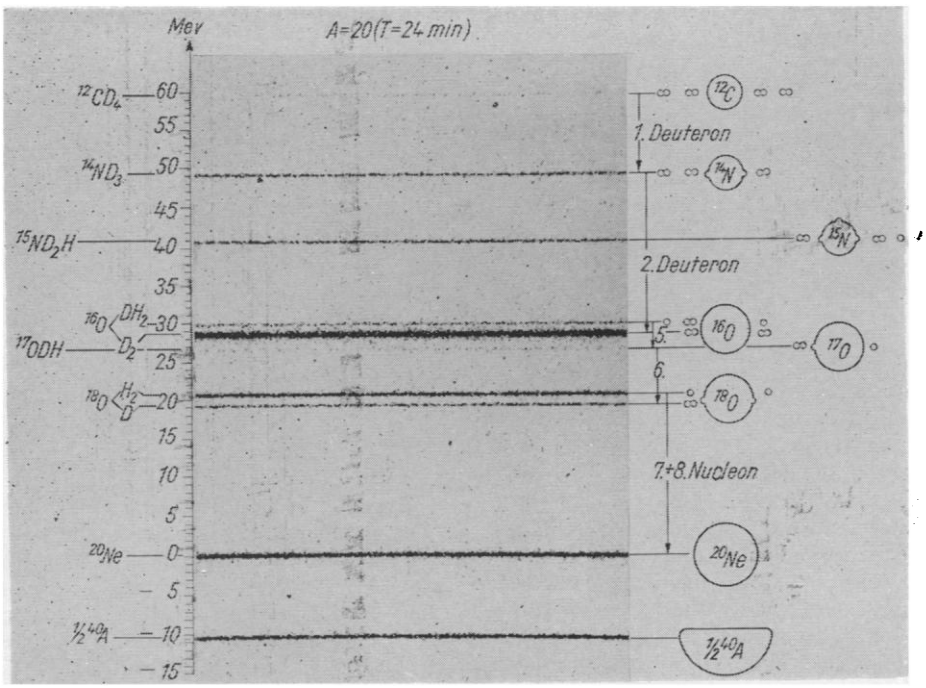

Fig. 8. Binding energies of successive deuterons and nucleons in the sequence that leads from ^{12}C over ^{16}O to ^{20}Ne.

the binding energy of the proton following ^{13}C, which leads to ^{14}N.

The subshell resulting from the building in of two neutrons with opposite spin is apparently better defined. The difference ($^{18}OH_2 - ^{20}Ne$) yields directly a value of 20.8 Mev for the binding energy of the now following proton pair. It is 1.1 Mev smaller than the corresponding energy of the nucleon pair, which leads from ^{14}N to ^{16}O. One recognizes the loosening up that is brought about by the electrostatic repulsion of the two protons. Hence, the sum of the binding energies of the four nucleons, which leads from ^{16}O to ^{20}Ne, is 2.4 Mev smaller than the one for the first four nucleons which follow the ^{12}C nucleus. Whereas the ^{12}C nucleus fuses the first alpha particle (thereby becoming ^{16}O) with a binding energy of 7.15 Mev, the ^{16}O nucleus is able to bind the next alpha particle (leading to ^{20}Ne) with an energy of only 4.75 Mev.

In the energy-level scheme of the closed shells of four the ^{16}O nucleus is therefore particularly low-lying. In this nucleus another shell of a different type is closed, which is characterized further by the magic number 8. The number of protons, as well as the number of neutrons in the ^{16}O nucleus, is 8.

It is also possible that lines below the "ground state" occur in our level scheme. For example, in Fig. 8, the line produced by the doubly charged ^{40}A ions shows that in one-half of a ^{40}A nucleus the nucleons are packed still more densely with an energy of 10.5 Mev, than they are in the ^{20}Ne nucleus. In the ^{40}A nucleus the binding energy per nucleon is therefore on the average about 0.5 Mev larger than it is in the ^{20}Ne nucleus.

In view of some small but definite discrepancies in the masses of some substandards with respect to which most nuclidic weights are measured, and on the accurate value of which in relation to the standard ^{16}O various groups of physicists have not been able to agree upon, I would like to mention that we have tried particularly to increase the accuracy of these measurements. In a paper (*4*) just published we were able to measure such substandards with an error of 1 in 3 million, and we hope with photographs, some of which are shown here, to reach an accuracy of 1 in 10 million.

References and Notes

1. This article is based on a lecture given at the meeting of the German Bunsen Society for Physical Chemistry in Bayreuth, 28 May 1954. It was first published in *Z. Elektrochem.* **58**, (1954) and is reprinted here by permission. I am indebted to Mrs. A. M. Akeley and H. Paul, Purdue University, for the translation of the manuscript.
2. A detailed description of the double focusing mass spectrograph which we are now using and with which the photographs shown here were made will appear shortly in the *Z. Naturforsch.* I would like gratefully to acknowledge the enthusiastic help of my two young collaborators, R. Bieri and F. Everling, who are largely responsible for the technical improvements of the apparatus and the taking of the photographs.
3. I would like to express my thanks to K. Clusius for the first two; the latter we received from Harwell.
4. J. Mattauch and R. Bieri, *Z. Naturforsch.* **9a**, 303 (1954).