

**Chemistry.** — "*The Mass-Ratio of Isotopes in Chemical Elements.*"

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Since ASTON, by an ingenious modification of J. J. THOMSON'S method of positive rays, was able to prove the complex nature also of a number of non-radioactive elements, — the question has obtained renewed interest for the chemist, as to what significance should really be attributed to the ordinary atomic weights of the elements, as finally determined after the accurate investigations during more than a century. More particularly there is reason to ask, whether the atomic weights of the common, non-radioactive elements can any longer be considered as real constants or not? For in the case of some elements, isotopes of certain products of radioactive desintegration, it has been proved on the one side, that their apparent atomic weights were different, according to the special nature of the radioactive minerals from which they were separated, just because of the unequal ratio in which in each case the isotopous desintegrations-products, no longer separable by chemical means, were present in their mixture. On the other hand it followed from ASTON'S investigations, that the greater number of the ordinary elements studied by him, consists of isotopous components, the atomic weights of these being represented by whole numbers, if calculated with respect to that of oxygenium = 16. The values of the observed atomic weights of the chemical elements, — which in general deviate from whole numbers, — must, therefore, depend on the special ratio in which the isotopous components are present in the mixture, formerly considered to be homogeneous in nature. If under different circumstances this mixing ratio should also appear to be variable, then the atomic weights of the common elements, as determined in the usual way, could no longer be considered as being true constants of nature.

However, experiments up till now seem to indicate, that no such variability of the ordinary atomic weights can be observed. At this moment a great number of values of atomic weights are accurately known as a result of the very exact atomic weight-determinations of the last thirty years. Within the limits of the inevitable experimental

errors, those values appeared always to be identical. From this fact it seems safe to draw the conclusion, that the mixing ratios of the isotopous components with their whole-numbered atomic weights, are in ordinary elements under all circumstances the same and invariable. It is, however, not so easy to answer the question *why* this is the case. For it appears that it is possible to consider this question from more than a single point of view.

It may be supposed, — and this is the oldest point of view, accepted originally by physicists, — that the atomic weights appear therefore always to have the same values, because the materials studied in reality represent essentially the same objects. For in the course of the enormous geological ages, measuring hundreds of millions of years, that have elapsed since the genetic process of the elements, — all matter in the solar system has been so thoroughly mixed up, that e.g. all local differences which eventually might have once existed on the earth, have long since become effaced. Each investigated element, therefore, independent of its locality on earth, may thus be considered as being in all cases taken from the same and completely homogeneous main store.

On the contrary, also another point of view may be adopted: it may be supposed that a more intrinsic cause, connected with the nature of the atoms themselves, is responsible for the apparently constant ratio of the isotopous kinds of atoms in the so-called chemical elements, instead of being that cause attributed solely to the homogenizing influence of external circumstances on matter. The case might be, that the said constant ratio might depend on the degree of relative mechanical or electromagnetic stability of the very complex moving systems we call atoms, it being in a certain way the expression of the equilibrium-ratio determined by the relative degree of probability of the different atomic systems, as it existed at the moment when those systems took their origin simultaneously from the chaotic mass of irregularly moving protons and electrons by the primordial process of differentiation. If — let us say — *two* kinds of atomic systems with equal nuclear charges, but different nuclear masses, originated in that process, the number of atoms of each kind then produced would evidently depend on the relative probability or stability of each of them under those circumstances. The fact that in our common elements the mass-ratio of two isotopous components is often strongly shifted towards one of them, so that the atoms of that kind are present in the mixture in a preponderant quantity, — may be considered as an indication, that really their probability during the genetic process was not altogether the same.

Moreover, it is probable that external circumstances only played a secondary rôle during that process: at least experience teaches us, that also in other respects the variation of external parameters does not influence appreciably the special rate or character of such intra-atomistic phenomena.

Finally a third supposition has been made by some authors, namely, that the mixtures of isotopes we call „elements”, are residues or products of former radioactive desintegrations, just as this is the case for the “lead”-species of variable atomic weights met with in radioactive minerals; be it that these mixtures are really inactive final products of such desintegration-processes, which are very slowly transformed with accompanying radiation, or without such radiation; or that they are mixtures whose radioactivity cannot be demonstrated by our actual means. In the last case it might be possible that, as in the case of the “lead”-species mentioned, a difference in atomic weight could be found, dependent on the former history of these products. We could e.g. imagine, that differences in atomic weight were demonstrable by sufficiently exact methods, if the elements considered originated from different geological formations or from different localities on earth.

In later years the opinion with respect to the problem under consideration has gradually gone in the direction of the second supposition mentioned above, i.e. of that, according to which an intrinsic, statistical cause should exist which determined the special ratio of the isotopes, as found in the common elements; this cause being then related to certain factors of stability of the different atomic systems. This view would be strongly sustained, if it were possible to prove the identity of the ratio of the isotopous components also for an element of cosmic origin, whose generative circumstances very probably have been quite different than for the same element on earth.

The only way for such an investigation is afforded by the study of that cosmic material which reaches the earth as *meteorites*. Leaving for the moment aside the question, whether these meteorites always formed a part of the solar system or if they came from the outside of it, — it must, however, appear certain, that the generative circumstances of those cosmic masses were completely different from those of terrestrial aggregates of analogous chemical constitution. More especially is this the case with that class of meteorites, which ordinarily is discerned as that of the *aerolites* or *chondrites*. For the greater part they consist of compounds of *siliciumdioxide* with numerous bases, as e.g. those of *calcium*, *magnesium*, *iron*, *alu-*

*minium*, etc. They occur in aggregates which, as follows from their petrographic character, originated under circumstances totally different from such as were present in the formation of terrestrial rocks of analogous chemical constitution. From ASTON's work it follows, that *silicium* consists of *two* isotopes, whose atomic weights are **28** and **29**; the atomic weight of ordinary *silicium* being higher than **28** and lower than **29**, indicates a mixture of definite ratio of both components, which appears as a chemically homogeneous substance.

It has been our purpose to determine with the utmost accuracy the ratio of both isotopes in terrestrial *silicium* of different origin and to compare it with the same ratio in *silicium* of cosmic origin. Perhaps it would be possible in this way to state a difference in the ratio of both isotopes in these different kinds of *siliciumdioxide*. In accordance with this we investigated in the way described further on the *silicium* from a number of terrestrial minerals of very different local and geological origin and afterwards from a number of chondrites of different petrographical character and of widely deviating times and localities of fall, so that the probability was increased as much as possible to find amongst those objects material of different cosmic origin. The results of these measurements, executed with the highest accuracy momentarily attainable were all obtained in this laboratory and will be published within a short time in all details in the dissertation of Mr. DIJKSTRA. Therefore a concise review of the method applied and the chief results obtained must suffice here.

The method which in the first place presents itself here would be a direct atomic weight-determination of the *silicium* separated from all these silicates. However, this method could not be followed here, because the atomic weight of the element *silicium* is one of the least accurately determined values of this kind. The data available in literature oscillate between 28,06 and 28,39, the cause of which are the extraordinary difficulties met with in the exact atomic weight-determination as a consequence of the particular properties of the *silicium*-compounds suited for this purpose. But even the best and most accurate atomic weight-determinations would not afford a degree of accuracy great enough for the purpose desired in this case, so that even then it would not be allowable to deduce with any certainty the very small deviations of the ratio of the isotopes in mixtures of them, where the atomic weights of both components do not differ more than a single unit. Disregarding here the possibility of the existence of a third *silicium*-isotope (with a mass = 30), the measured atomic weight would correspond with a mixture con-

taining 6% *Si* of the heavier and 94% of the other kind, while the atomic weight-determination of 28,39 would on the other hand correspond with a mixture of 39% *Si* of the mass 29 and 61% *Si* of the mass 28. From this it becomes evident that an error in the atomic weight-determination of only 0,1% would cause an apparent shift of the ratio considered of no less than 3%.

Therefore we have used a fact which enables us to reach a much greater accuracy. The chemical properties of isotopous elements, only depending on the outer electrons, are rigorously identical. If a certain ratio of them is, therefore, present in the mixture we call the element *silicium*, it will be clear that, independent of what chemical reactions are executed with it, that ratio will remain unchanged in all derivatives prepared from it. Moreover, the specific weight of isotopous elements is directly proportional to their atomic weight. The fact is most probable, because they occupy the same place in the periodical system of elements, having, therefore, identical atomic volumes, and from this the proportionality between atomic and specific weights follows immediately. That this view is a sound one was proved by experiment by the accurate determination of SODDY and HYMAN<sup>1)</sup> with *thorite*-, „lead” and by those of RICHARDS<sup>2)</sup> with isotopous mixtures of „lead” from different radioactive minerals: both atomic and specific weights of these mixtures were determined and a rigorous proportionality between both constants was found in all cases. The same is true also for the *derivatives* of such elements, as was demonstrated for the *nitrates* of these „lead”-species by FAJANS and LEMBERT<sup>3)</sup>.

Every real deviation of the specific weight of isotopes or of that of the compounds prepared from it, must thus necessarily correspond with a real shift in the ratio of the isotopous components in the mixture. Much more exactly than by a direct atomic weight-determination, the ratio considered can be determined by comparison of the specific weights of the same derivative of *silicium*, if measured with the highest attainable accuracy.

To fix the choice of a compound suited for this purpose, the question arose to find a perfectly stable liquid substance, which

<sup>1)</sup> F. SODDY and H. HYMAN. *Trans. Chem. Soc.* **105**, 1402, (1914). The specific weight of *thorite*-„lead” was 0,26% higher than that of ordinary lead; but at the same time the atomic weight as determined by SODDY (207,69) and by HÖNIGSCHMID (207,77) was in agreement with the mentioned supposition, which involves in this case a value of 207,74.

<sup>2)</sup> TH. W. RICHARDS, *Pres. Address Amer. Assoc. Adv. of Science.*

<sup>3)</sup> K. FAJANS and M. LEMBERT, *Zeits. f. anorg. Chem.*, **95**, 297, (1916).

could be obtained in a completely pure state and which possesses such properties, e. g. of unattackability, that it might be used for these measurements without objections. Finally we chose *tetra-ethyl-silicane*:  $\text{Si}(\text{C}_2\text{H}_5)_4$ , a colourless liquid with a boilingpoint of  $154^\circ$  C., which in many respects shows a character like that of the *paraffines*. It is highly stable and may e. g. be shaken during many days with concentrated sulphuric acid, without showing any trace of decomposition. Moreover, it follows from ASTON's experiments, that *carbon* and *hydrogen* are both "full-bred" elements, without isotopous components, so that the whole effect eventually to be found must be wholly ascribed to a change in the ratio of the *silicium*-components alone. We preferred this compound to the corresponding *tetramethyl-silicane*, because this last has a boilingpoint some  $120^\circ$  lower than the first, so that it is much more volatile and might, therefore, more easily cause errors in the pycnometric determinations, while on the other hand a fractionating within a few hundreds of a degree may be executed with less difficulty in the case of the higher boiling liquid.

The preparation of the *tetra-ethyl-silicane* from the different kinds of *siliciumdioxide* was made as follows. The *siliciumdioxide* required was separated from the minerals and chondrites by melting the finely powdered silicates in platinum crucibles, carefully and in small quantities to avoid explosions in the case of the chondrites, with an excess of pure *sodiumcarbonate* and *sodiumnitrate*, heating the product with *hydrochloric acid* and repeating this treatment several times with the filtered, washed and dried *siliciumdioxide*. Particularly in the case of the chondrites, this operation must be executed with great care, because they contain *schreibersite* (*ironphosphide*), *pyrrhotine*, *troilite* and other *sulphides*; the platinum-crucibles are strongly attacked therefore in these cases. Finally the *siliciumdioxide* was obtained in this way in the form of a pure white, voluminous powder, which was converted into the desired compound in the following way.

The *dioxide* was transformed in a silver dish by means of a solution of *ammonium-fluoride* carefully freed from all *siliciumdioxide*, into *ammoniumhexafluosilicate*:  $(\text{NH}_4)_2\text{SiF}_6$ , by addition of strong *hydrochloric acid*. This salt was converted into the corresponding *barium-salt*, which on heating is decomposed into *barium-fluoride* and *siliciumtetrafluoride*:  $\text{SiF}_4$ . This gas was led in a perfectly pure and dry state into a perfectly dry ethereal solution of a great excess of GRIGNARD's reagents, in which *magnesium-ethyl-bromide* was the active substance. All possible care was taken to prevent any contamination of the product; finally the substance was

obtained in a perfectly pure state as a colourless liquid, boiling at a pressure of 760 mM. at  $154^{\circ},95$  C. Analysis gave:  $66,48\%$  C and  $14,54\%$  H; calculated:  $66,49\%$  C and  $13,97\%$  H. A molecular weight-determination showed, that the molecular weight really corresponded with that of  $Si(C_2H_5)_4$ .

In this way *twelve* different preparations were obtained: *six* from *terrestrial* silicates of strongly deviating origin, namely from *quartz-sand* from *East Groningen*, from a *German* and a *North American* locality, further from *analcite* (*Seisser Alp*), from *leucite* (*Tavolato* near *Frascati*) and from *chlorite* (*Pfitschtal*). *Analcite* is a zeolithic silicate, *leucite* a volcanic product, *chlorite* a silicate of metamorphic origin.

Six other preparations were obtained from *siliciumdioxide* of *cosmic* origin: from an *aërolite* of *Alfanello* in *Italy* ( $16^{\text{th}}$  february 1883), of *Soko Banya* in *Servia* ( $13^{\text{th}}$  october 1872), of *Holbrook* in *Arizona* ( $19^{\text{th}}$  july 1912), of *Ochansk* in *Russia* (*Perm*) ( $30^{\text{th}}$  august 1887), of *Bjurböle* in *Finnland* ( $12^{\text{th}}$  march 1899) and of *Kesen* in *Japan* ( $13^{\text{th}}$  june 1850). Of these chondrites that from *Soko Banya* is a *peridotite-enstatite-chondrite* with fully  $40\%$   $SiO_2$ ; that from *Ochansk* a *hypersthene-olivine-chondrite* with  $37\%$   $SiO_2$ ; that from *Holbrook* a typical *enstatite-chondrite* with  $30,8\%$   $SiO_2$ ; the chondrite of *Bjurböle* had  $41\%$   $SiO_2$ . Besides *pyrrhotine*, *troilite* and *schreibersite*, small granules of *iron-nickel* and *chromite* were found; some chondrites contained also traces of *copper*, probably in the form of sulphur-compounds. In Fig. 1—3 some photographs are reproduced of microscopical sections in ordinary and polarized light; the black inclusions consist of *troilite*, *chromite* and *iron-nickel*. The powder of these chondrites is pale ash-grey to greyish-brown; they possess a trass-like, finely granular structure.

The pycnometer used had a volume of about  $3\text{ cm}^3$ . and was of the wellknown OSTWALD-SPRENGEL-type, slightly modified and made of a special kind of hard glass for the purpose of preventing a change of the glass-surface by the water of the thermostate. The temperature of the thermostate could be kept constant within  $0^{\circ}.01$  C. and was perfectly constant (within  $0^{\circ}.001$  C.) during the short time, that the pycnometer was immersed. To prevent as much as possible the correction for the mercury-column above the water-surface, the thermometer was circumflowed totally by the water of the thermostate and so read, that no incertitude by parallaxis was introduced. All weighings were executed by means of a micro-balance, which was enclosed in a box of red copper to protect it against temperature-radiations. Instead of by the ordinary mirror-readings, the

oscillations of its beam were measured at a considerable distance by means of a telescope and a movable lighting spot, obtained by means of a reflected light-beam in a small mirror fixed in the middle of the beam of the instrument and a prism-arrangement. All weights were calibrated, as in atomic weight-determinations and reduced on the vacuum; the same was the case with all weighings, while the corrections for the upward pressure were made as small as possible by applying a counter-weight of about the same volume as the pycnometer had. No measurements were made before the pycnometer had hung in the balance-box for three quarters of an hour, when it completely had accepted the temperature of its environment. All radiations from the human body were prevented in this way, because all readings were made at a considerable distance from the instrument. Arrangements were made to prevent the occurrence of electrical charges at the agate supports and at the walls of the balance which might cause a shift of the zero-point, by placing some radioactive *thorium-carbonate* in the box of the instrument; by preliminary experiments all corrections were moreover exactly determined with respect to the inequality of the balance-beams, the changes of atmospheric pressure, of temperature, etc. To give an impression of the high degree of reproducibility of these measurements acquired, it may be mentioned, that in subsequent weighings of a mass of about totally 10,5 grammes, identical results were obtained within a few unities of the *sixth* decimals, after all necessary corrections had been made: hundredth parts of milligramms can thus be determined with perfect certainty and only the thousands parts possess less significance. Originally some deviations occurred by the filling of the pycnometer as a consequence of capillar and other influences; after a modified construction of the apparatus and another kind of glass being chosen, this difficulty also was finally overcome.

By far the greatest difficulties were met with during this investigation in the purification of the different preparations. In the GRIGNARD-reaction a number of by-products take their origin, chiefly some *ethyl*-derivatives and among them *tri-ethylsilicol* ( $C_2H_6$ ),  $SiOH$ , the boilingpoint of which differs only slightly from that of *tetra-ethyl-silicane*, being therefore only with great difficulty separable from it by fractional distillation. Complete purification appeared only possible by repeating the GRIGNARD-reaction several times, by reduction of still present *silicol* and decomposition of the also formed *triethylsilicol-ethylether* by heating with *hydrojodic acid* during several days, shaking for a long time with concentrated *sulphuric acid* and



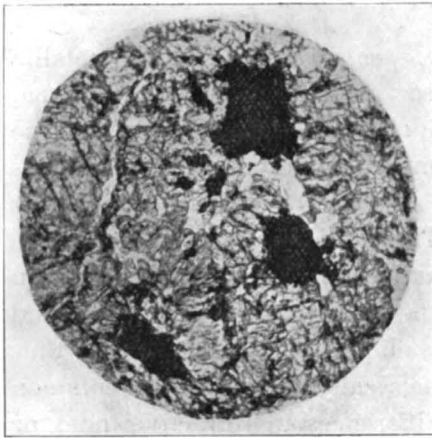


Fig. 1a. Chondrite of *Alfanello*  
in ordinary light.

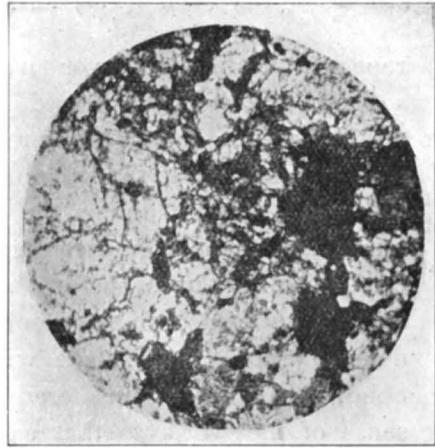


Fig. 1b. Chondrite of *Alfanello*.  
between crossed nicols.



Fig. 2a. Chondrite of *Soko Banya*  
in ordinary light.



Fig. 2b. Chondrite of *Soko Banya*  
between crossed nicols.

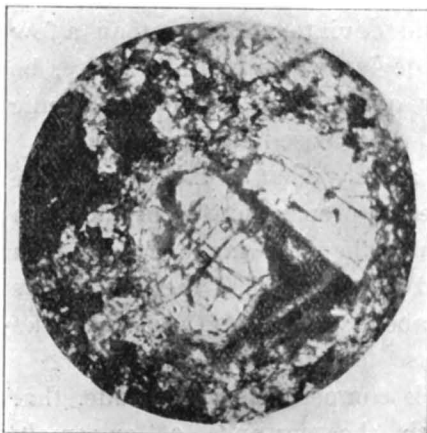


Fig. 3a. Chondrite of *Ochansk*  
in ordinary light.

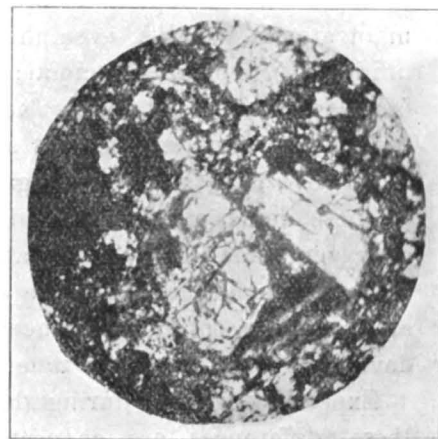


Fig. 3b. Chondrite of *Ochansk*  
between crossed nicols.

repeated distillation over *phosphorus-pentoxide* and fresh metallic *sodium*. Finally, the liquid obtained was subjected to fractional distillation under constant pressure and, after calculation of the true boiling-point from the vapour-tension-curve, only that fraction was separated, which under the given pressure boiled within a few hundredths of a degree centigrade. The liquid was not considered to be sufficiently pure, before no deviations of the specific weight in subsequent measurements could be stated any longer. Then the final determinations were made with this liquid. The preparations thus obtained might be considered as "chemically pure" in the common sense of the word, so that e.g. no differences in refractive index or in chemical behaviour could be stated any more. The numerous details of the technical side of these experiments may be seen from the larger publication mentioned above.

In the table on page 403 the results of these measurements are reviewed.

From this it is evident that the specific weights at 20° C. of the *tetra-ethyl-silicane* of *terrestrial* origin, independent of the fact whether the *siliciumdioxide* used came from *quartz-sand* of different localities or from silicates of zeolithic, volcanic or metamorphic character, do not differ from each other more than 0,006 ‰ of the main value: 0,76734 ± 0,00005; the deviations are irregular to both sides. For the *cosmic* preparations the main value is: 0,76735 ± 0,00003; the deviations are not greater than ± 0,005 ‰. The differences between the determined values with terrestrial and cosmic material are not greater than about 0,002 to 0,003 ‰.

The values indicated with  $\Delta$  are the variations in main atomic weight, as they may be calculated from the observed specific weights, supposing that their differences were really caused by a different ratio of the isotopous components of *silicium*. It appears that the main atomic weights, even then, could deviate no more than a few unities of the *third* decimal; these deviations would, therefore, be far beyond the attainable accuracy of the best method of atomic weight-determination, as was already emphasized in the above.

However, it must now again be asked, what is the significance of these deviations found in the different preparations? For, although only very small, they are real, because they are much greater than the experimental error of the method followed. The method allows an accuracy of measurement of about 0,0004 ‰, so that the deviations mentioned are in every case ten times as great.

Experience gained during this work compels us to conclude, that these differences are connected with the presence of extremely

minute traces of by-products, admixed with the preparations of *tetra-ethyl-silicane*: traces so small, that they cannot be indicated any longer by any reagent, and that they cannot influence in any way the chemical character of the different substances.

To get more information about this and greater certainty, we

A. Terrestrial material:	Boilingpoints of fraction:	Theoret. boiling-point:	Weight of Pycnometer-	Specific <sup>1)</sup> weight at 20° C.	(n-n'):	Δ
Netherlands (Groningen)	$p=748,8; 154^{\circ},10-154^{\circ},15$	154°,1	2,27760	0,76730	+0,000026	-0,0095
Germany	$p=756,6; 154^{\circ},50-154^{\circ},54$	154°,5	2,27766	0,76732	+0,000024	-0,0057
N. America (Penns.)	$p=765,8; 154^{\circ},84-155^{\circ},0$	155°,0	2,27761	0,76730	+0,000082	-0,0089
Analcite (Seisser Alp)	$p=749,0; 154^{\circ},16-154^{\circ},20$	154°,1	2,27790	0,76740	-0,000005	+0,0095
Leucite (Tavolato, Frascati)	$p=765,5; 154^{\circ},98-155^{\circ},06$	155°,0	2,27784	0,76738	-0,000002	+0,0057
Chlorite (Pfitschtal)	$p=756,2; 154^{\circ},46-154^{\circ},60$	154°,5	2,27778	0,76736	+0,000005	+0,0019
<b>B. Cosmic material:</b>						
Chondrite of Alfanello (Italia) 16. 10. 1883.	$p=760,8; 154^{\circ},62-154^{\circ},70$	154°,7	2,27774	0,76735	+0,000018	-0,0006
Chondrite of Soko Banya (Servia) 13. 10. 1872.	$p=763,4; 154^{\circ},84-154^{\circ},90$	154°,9	2,27776	0,76735	+0,000012	+0,0006
Chondrite of Holbrook (Arizona) 19. 7. 1912.	$p=759,8; 154^{\circ},66-154^{\circ},70$	154°,7	2,27776	0,76735	+0,000013	+0,0006
Chondrite of Ochansk (Perm.) 30. 8. 1887.	$p=760,6; 154^{\circ},65-154^{\circ},68$	154°,7	2,27784	0,76738	—	+0,0057
Chondrite of Björbole (Finland) 12. 3. 1899.	$p=765,2; 154^{\circ},92-154^{\circ},96$	154°,9	2,27775	0,76735	+0,000002	0
Chondrite of Kesen (Japan)	$p=753,9; 154^{\circ},35-154^{\circ},50$	154°,4	2,27776	0,76735	+0,000001	+0,0006

<sup>1)</sup> The real temperature was 19°,93<sup>5</sup> C.; in abbreviation 20° C. is written.

have determined *the differences of the refractive indices for sodium-light* between the different preparations by means of the interferometer of LOEWE, as constructed by ZEISS. If the deviations found were really caused by a variation of the mixing ratio of the *silicium*-isotopes, there could *not* be any difference between the refractive indices, because the optical properties of isotopous elements for wavelengths within the limits of the visible spectrum are identical, as being again only a function of the outer electrons of the atoms. But if these deviations were connected with the presence of minimal traces of impurities, there would be a good chance, that the interferometric method would really reveal such differences in the refractive indices. In that case, however, a rigorous proportionality between the deviations of the specific weights and those of the refractive indices could, of course, not be expected: there are, as we have seen, several kinds of impurities possible here, and they will be simultaneously present in variable and unknown quantities in each of the preparations. At the best there could be expected a kind of rough anti-parallelism <sup>1)</sup> between both series of numbers.

The measurements were executed in such a way, that as the liquid of comparison in all cases, the *tetra-ethyl-silicane* from the meteorite of *Ochansk* was always taken in one of the two vessels of the chamber of the instrument. The deviations of the refractive indices of the other preparations with respect to the liquid mentioned, are given in the Table under  $(n-n')$ ; the division of the drum of the micrometer is such, that on using this special chamber, each division corresponds to two units of the *sixth* decimal of the refractive index.

If now the *tetra-ethyl-silicane*-preparations were arranged in the same way as in the Table and the values of the specific weights or of the numbers  $(n-n')$  were taken as ordinates in a graphic figure (Fig. 4), two broken lines were obtained of the form represented therein; the numbers  $(n-n')$  are taken with opposite algebraic signs. Although evidently a strong proportionality is not present, nobody can doubt any longer the striking parallelism between both series of values. From this it must be considered as highly probable, that the deviations of the specific weights of the *tetra-ethyl-silicane*-preparations have no real significance, as far as regards the eventual difference in ratio of the *silicium*-isotopes in them: for in that case the values  $(n-n')$  must necessarily be all equal to *zero*. Within the limits of the

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<sup>1)</sup> *Anti-parallelism*, because the specific weight of the preparations is lower, as they are purer. Therefore the algebraic signs of  $(n-n')$  are inversed in Fig. 4.

highest accuracy at this moment attainable, the specific weights, and, therefore, *the said ratios of the isotopes, must consequently be*

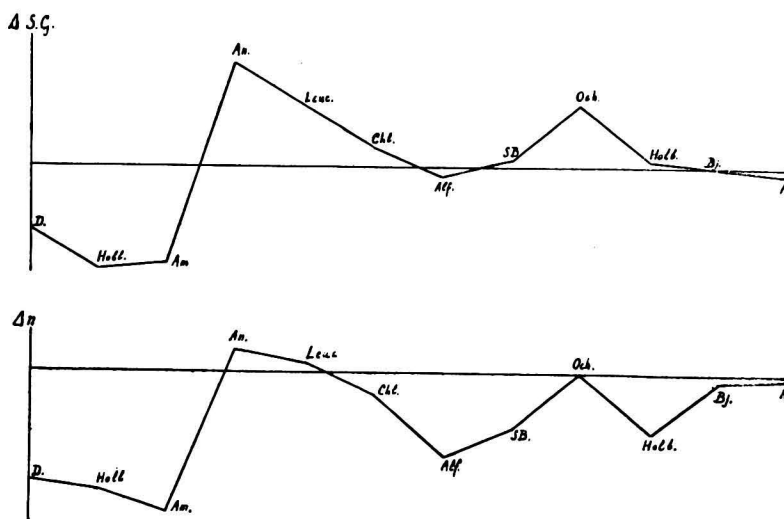


Fig. 4.

*identical as well in the case of the terrestrial as of the cosmic silicon-species.*

In connection with these results attention may finally be drawn to the analogous conclusions, to which other investigators before us have come in this respect, — even although to these results, with a single exception perhaps, no such high degree of accuracy can be attributed as to ours, as a consequence of the methods applied.

BAXTER and THORVALDSEN, HOOVER, PARSONS and HILTON <sup>1)</sup> found the atomic weights of *iron* and of *nickel* from the *Cumpas*-meteorite identical with the atomic weights of the terrestrial elements within the limits of experimental errors. I. CURIE <sup>2)</sup> compared the corresponding value for *chlorine* from seawater, from a Canadian *sodalite* and from *apatite* by weighing of *silverchloride*; GLEDITSCH, SAMDAHL <sup>3)</sup> and DORENFELDT <sup>4)</sup> proved the same identity for *chlorine* from a Norwegian and another specimen of *apatite*; BRÖNSTED and VON

<sup>1)</sup> G. P. BAXTER and T. THORVALDSEN, Journ. Am. Chem. Soc., **33**, 337, (1911).

G. P. BAXTER and C. R. HOOVER, Journ. Amer. Chem. Soc., **34**, 1657, (1912).

G. P. BAXTER and L. W. PARSONS, Journ. Amer. Chem. Soc., **43**, 507, (1921).

G. P. BAXTER and F. A. HILTON, Journ. Amer. Chem. Soc., **45**, 694, (1923).

<sup>2)</sup> I. CURIE, Compt. rend. Paris, **172** 1025, (1921).

<sup>3)</sup> E. GLEDITSCH et M. B. SAMDAHL, Compt. rend. Paris, **174**, 746, (1922).

<sup>4)</sup> M. DORENFELDT, Journ. Amer. Chem. Soc., **45**, 1577, (1923).

HEVESY <sup>1)</sup> found by accurate measurement of the specific weight of *mercury* of nine different localities values, which did not differ 0,1 %; RICHARDS <sup>2)</sup> did neither succeed in finding a difference in the atomic weights of *copper*, and of the *calcium* of *calciumcarbonate* and the *chlorine* of *sodiumchloride* from different localities; MONRO <sup>3)</sup> also came to the same conclusion in his measurements of *borium* (titration-method). On the contrary, MUZAFFAR <sup>4)</sup> mentioned differences of 1 to 1,5 % in the atomic weight of *antimony* from different localities (titration-method). But these experiments do not give the impression of sufficient accuracy, so that it will be best to wait for further determinations, before a final judgement is given.

The investigation described in the above will convince us of the fact, that the higher the degree of accuracy of the method followed and the more vigorous the critical discussion of the deviations eventually met with, the greater the chance to find, that such deviations are of no immediate significance with respect to the problem involved. Independent of their origin, the chemical elements seem really always to have a *constant* ratio of their isotopous components, which ratio is, therefore, *characteristic* for each of them. But then the character of true *constants* of nature must indeed be attributed to the atomic weights of the elements, as ordinarily determined.

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*Groningen, Netherlands, April 1924.*

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<sup>2)</sup> TH. W. RICHARDS, Proceed. Amer. Acad. of Arts and Sc., **23**, 177, (1887); Journ. Amer. Chem. Soc., **24**, 374, (1902).

<sup>3)</sup> A. D. MONRO, Journ. Chem. Soc. London, **121**, 986, (1922).

<sup>4)</sup> S. D. MUZAFFAR, Journ. Amer. Chem. Soc., **45**, 2009, (1923).