

abode, the universe. let him learn to estimate aright the earth, its kingdoms, its cities and himself. What is man in the infinite?

"But to show him another marvel, no less astonishing, let him examine the minutest things he knows. Let him consider a mite and note the tiny body composed of parts incomparably more minute; the limbs with joints, the veins in the limbs, blood in the veins, humours in the blood, drops in the humours, and vapours in the drops. Let him again divide these parts, exhausting his powers of imagination, and he may think he has arrived at the most extreme diminutive in nature. Then I will open before him a new abyss. I will depict for him not only the visible universe, but all the immensity of nature imaginable in the enclosing envelope of this minute atom. Let him see therein an infinity of universes, each with its firmament, planets and earth in the same proportion as in the visible world. In each earth animals, down to the midget existences that show him all that he has already seen in the first. However many he may see he will find in all the same unending, unresting purpose, and he will lose himself in all these

marvels, as wonderful in their minuteness as the others in their immensity, for who will not be amazed to realise that our human body just now perceived to be but an imperceptible atom in an insignificant planet of the universe, now becomes a colossus, a world, a vast whole with regard to the nothingness into which we cannot penetrate?

"Whoever sees himself in this way will be terrified of himself, and, considering how he is upheld in the material substance nature has given him between the two abysses of the infinite and nothing, he will tremble at the sight of such marvels; and I think that as his curiosity changes into wonder he will be more disposed to contemplate them in silence than to presume to investigate them.

"For after all, what is man in nature? A nothing compared with the infinite; a whole with regard to nothing, a mean between nothing and everything. Infinitely far removed as he is from understanding either extreme, to him the end of all things and their beginnings are hidden in a baffling, impenetrable mystery; he can see neither the nothingness whence he was taken, nor the infinity in which he is engulfed."

### The Theory of Valency: Development and Problems.

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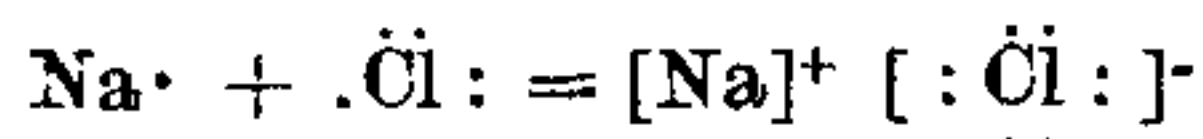
**D**URING the last century chemistry built up the system of molecular structures as we know it to-day. Firstly and mainly it is characterised by the phenomenon of "saturation of valency". The chemical forces of the atoms were described by valency bonds which represented the number of valencies of each atom, two of these bonds were able to combine two atoms in chemical union, and molecules with unsaturated valencies existed only under abnormal conditions as in free radicals or in many unsaturated molecules such as  $\text{BeF}$ ,  $\text{BeH}$ ,  $\text{AlH}$ ,  $\text{CH}$ , etc., which the physicist observes in the electric arc or in the discharge tube. It was of course necessary to assume that many atoms possessed different valency numbers but modern atomic physics has furnished such a simple and obvious explanation of this phenomenon, that we do not see any difficulty in this second assumption to-day. The simple rules worked out by Chemistry during the last century meet the requirements of all primary molecules.

Chemistry, however, was unable to give any explanation of the mechanism of chemical combination in either case, and had to be content to introduce names only for the unexplained chemical forces, such as affinity, forces of valency and so on, which could not be identified with those physical forces known at the time. Whereas Chemistry gave us a full answer to the question, *which* chemical union occurs if atoms approach each other, the question *why* it occurs and why it occurs just in this way could only be successfully taken up a century later by modern atomic physics. From the moment onwards when Bohr's theory of the structure of atoms made its appearance in 1913, it was clear that it could be only a question of time until the explanations of the problems would be found. The development took place in two steps. During the reign of the pre-wavemechanical quantum theory of the atom, heteropolar molecules could be explained and many of their properties quantitatively calculated and also some preliminary



models of homopolar molecular structures could be developed. Wave-mechanics opened the second epoch by explaining and (in some cases) directly calculating the forces which keep the neutral atoms of a homopolar molecule together and we may say that we have already advanced to a rather complete physical understanding of the principles involved in the chemical behaviour of atoms and molecules, even if many questions concerning details cannot fully be answered in the present moment.

In 1916 Kossel and Lewis simultaneously and independently put forward two theories, closely related to each other. The starting point of Kossel<sup>1</sup> was, that the number of outside electrons of free ions as they exist in solutions is identical with that of the nearest rare gas. Thus in sodium chloride the sodium atom has lost its eleventh electron and the positive sodium ion remains with the configuration of neon whereas the chlorine ion has gained this electron and has raised its electronic configuration to that of argon, which follows one step in the periodic system. Representing the outside electrons in the usual way by little dots (leaving the inner shells aside) we may represent this process by the following formula:



He therefore assumes that the electronic configuration of the rare gases is not only inert in a chemical sense but that also a peculiar physical stability is due to it, so much so that in the process of adding an electron to the neutral chlorine atom in excess of its nuclear charge, energy is liberated. This conception serves very well the demands of the heteropolar molecules which are made up of ions. The forces of the electrovalent link are therefore successfully identified with electrostatic forces between ions. Kossel uses it also in explaining homopolar linkages; it is, however, only fair to mention, that he considered this always as a first approximation only and hoped to overcome the obvious difficulties by considering the ions not as rigid balls but as capable of mutual polarisation.<sup>2</sup> This idea, so to speak the next approximation of Kossel's theory, was extended by

<sup>1</sup> W. Kossel, *Ann. d. Physik*, 1916, 49, 229. Cf. the monograph: "Valenzkräfte und Röntgenspektren" (Berlin, 1921).

<sup>2</sup> Cf. the discussion in *Zs. Elektrochem.*, Report annual meeting Bunsen Soc., 1928, pp. 24 and 60.

Fajans and his collaborators. In the meantime, however, Kossel was able to explain certain details of chemical behaviour without a special model of the homopolar linkage; thus the *maximal* valency number of nitrogen or phosphorus is 3 in combination with positive partners as in  $\text{PH}_3$ ,  $\text{NH}_3$ ,  $\text{AlN}$ , etc., because three electrons are missing for the completion of the configuration of the *following* rare gas, but it is 5 in combination with negative partners like in  $\text{N}_2\text{O}_5$  or  $\text{PCl}_5$  because these atoms possess 5 electrons more than the *preceding* rare gas. This agreement between valency number and the number of outside electrons shows that certain features even of the application of Kossel's theory to homopolar linkage cannot be overlooked in a definite formulation of a theory of valency.

Lewis<sup>3</sup> also started from the apparent stability of the eight-electron configuration of the higher rare gases. The main feature of the theory is, however, a special conception of the homopolar linkage which made it extremely useful in the hands of the organic chemist. From the fact, that nearly all stable homopolar molecules possess an even number of electrons, he concluded that a *pair* of electrons is responsible for each homopolar bond, without actual transfer of charge. The combination of two hydrogen atoms was written as follows:—



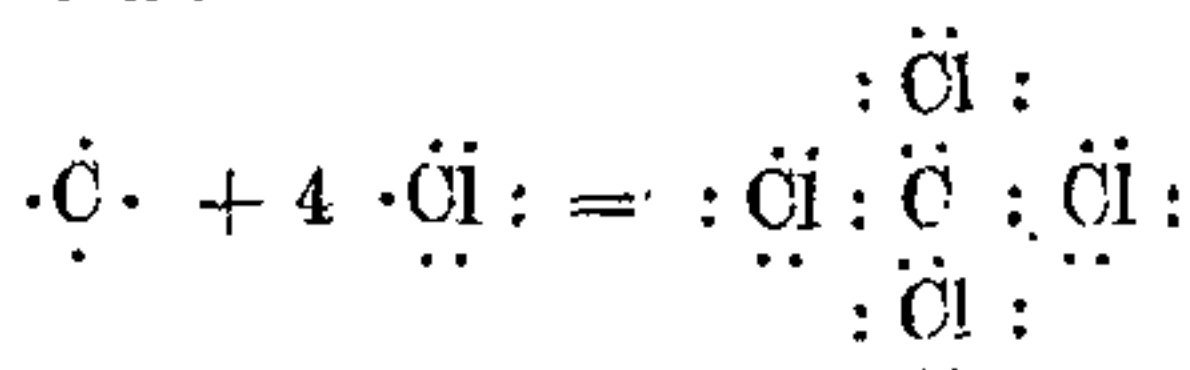
meaning, that in the molecule both electrons now are under the influence of the field of both nuclei. By this process of "sharing" the electrons, the system of two hydrogen atoms becomes more stable. Lewis believed magnetic forces to be the cause of the electron sharing. He could not know that quantum-mechanics would later reveal a then unknown force by which the "covalent" bond, as it was later called by Langmuir, is produced; but even this was not so far from the truth, since we know to-day that the spin of the electron plays a great part in chemical combination. The model of the hydrogen molecule, however, taken as such, was fully confirmed and substantiated by wave-mechanics.

In Lewis' model of the hydrogen molecule the electrons are redistributed in such a way, that each nucleus is related now to 2 electrons, that is, the number of electrons in the next inert gas, *i.e.*, helium. The combination

<sup>3</sup> G. N. Lewis, *J. Amer. Chem. Soc.*, 1916, 38, 762; *Proc. Nat. Ac. Amer.*, 1916, 2, 586. "Valence and the structure of atoms and molecules," 1923.



of four chlorine atoms with carbon to carbon tetrachloride takes place according to the formula :



Thus each nucleus is related to 8 electrons, that is, the number of the outside electrons, in the higher rare gases. He assumed, therefore, that the electrons tend also to form octet configurations in covalent molecules. Thus Lewis' theory contains two postulates: (1) the Duplet Rule which makes the formation of a pair of electrons responsible for each individual bond, and (2) the Octet Theory which assumes the tendency of octet formation. It should be clearly borne in mind, that these two postulates are independent of each other. This is important, because they are also completely inconsistent with each other. It is true that the system of organic chemistry can conveniently be described by the Octet Theory, but this is the automatic result of the prevailing tetravalency of carbon. If each chemical bond is represented by a pair of electrons, then each tetravalent atom will be surrounded by eight electrons in the molecule simply because four times two is eight. Organic chemistry deals only with a few atoms whose varying combination produces more than a hundred thousand molecules. In the periodic table we know, however, to-day 92 atoms, many of which possess different states of valency. Thus the tetravalent carbon atom is one only out of about two hundred cases of valency and every one of them is equally important; if we want to build up a theory of valency we have just to consider inorganic chemistry which represents a greater multiplicity and variety of chemical combinations. In the moment we leave alone the fourth group and the few cases, where the number of positive and negative valencies is equal and consider any other group of the periodic table, we see that either the Duplet Rule or the Octet Theory has to be abolished. Either the Octet Rule is strictly maintained, then the electrons have to be counted in such a way that their number when surrounding the central atom is increased on the left hand side of carbon in the periodic table and is decreased on its right hand side. This is done by introducing devices such

as the semipolar double bond and the singlet linkage. Or the Duplet Rule is strictly maintained, then the number of surrounding electrons is always double the number of valencies, i.e., mostly double the number of *p*-electrons or of *s* + *p*-electrons of the central atom and the significance of the Octet vanishes. Indeed from here onwards two different schools of thought have been developed on two different lines: the first theory is intimately connected with the names of Langmuir, Lowry, Prideaux, Sidgwick, Sugden a.o.,<sup>4</sup> the second was mainly developed by Grimm and Sommerfeld.<sup>5</sup> Incidentally Lewis himself attributed greater importance to the electron-pair-bond theory of linkage. Thus he states<sup>6</sup> "The striking prevalence of molecules in which each atom has its full quota of four electron pairs in the outermost shell has led Langmuir to attempt to make the Octet Rule absolute, and he even proposes an arithmetical equation to determine, in accordance with this rule, whether a given formula represents a possible chemical substance. I believe that in his enthusiasm for this idea he has been led into error, and that in calling the new theory the "Octet Theory" he over-emphasises what is after all but one feature of the new theory of valency. The rule of eight, in spite of its great importance, is less fundamental than the rule of two, which calls attention to the tendency for electrons to form pairs. The electron pair especially when it is held conjointly by two atoms, and thus constitutes the chemical bond, is the essential element in chemical structure."

Thus the pre-wave-mechanical, naive Quantum Theory gave us a complete understanding of the heteropolar chemical combination (taking the existence of electron affinity for granted) but was not able to explain that mutual interaction of electrons on which the electron affinity itself depends, and the homopolar linkage. Therefore, it was possible to interpret this phenomenon in different ways and two different schools of thought have been developed. One of them maintains the existence of particular stable electronic configurations around each atom

<sup>4</sup> Cf. Sidgwick, *Electronic Theory of Valency* (London, 1927).

<sup>5</sup> *Zs. f. Phys.*, 1926, 36, 38. Cf. Grimm's article in *Handb. d. Phys.*, 24. Lessheim and Samuel: "Die Valenzzahl, etc." (Berlin, 1927).

<sup>6</sup> "Valence etc." Chapter VIII. Cf. *J. Chem. Phys.*, 1933, 1, 23.



(Octet Theory) but has to resort to different varieties of non-electro-static linkage. The other one is a uniform theory which maintains the close relation between the number of valencies of an atom and the number of its electrons in its various outside groups and sub-groups and obtains stability in all cases in which the electrons of the central atom not taking part in the linkage form completed groups and sub-groups. We shall see later, that this difference of opinion leads even to-day to two different interpretations of the wave-mechanical treatment of the homopolar molecule, either being logical and self-contained, in such a way that we can decide between them only by comparing their results with experimental evidence.

If we are now going to consider the results achieved by wave-mechanics in recent years, we encounter the same difficulty with which every one who speaks or writes about quantum mechanics is faced. If we are coming to atomic dimensions, matter behaves unexpectedly different from its behaviour known to every one from the experience of daily life. We understand this behaviour in this sense, that we are able to describe it by valid mathematical formulæ and therefore wave-mechanics was able to explain covalent linkage between two atoms. If we take up, however, our leading question again and ask with which physical forces we have to identify now those "forces of valency," we are still at a loss to answer this question. It is—as a matter of principle—not possible to describe this behaviour by an analogy or a model based on daily experience or even in the language which was formed during hundred thousands of years by this experience of the macroscopic world and which, therefore, does not offer us either words or conceptions for such a description. In wave-mechanics, the Hamilton-Jacobi's equation of classical dynamics is replaced by a different equation, the so-called Schrödinger wave equation. Because the effects in the atomistic world are different from those in the microscopic world, they can be expressed only by a different method of calculating, which we call wave-mechanics and for which this equation is the foundation. In particular, covalent linkage is not due to an attraction of the atoms according to Coulomb's law but to a purely wave-mechanical effect which has no classical analogue but is somewhat similar to the classical resonance phenomenon. If the system of two atoms is degenerated in such a manner

that its energy value can be represented in two different ways—*e.g.*, by the exchange of electrons which are identical and whose exchange is therefore without an influence on the energy of the total system—this degeneracy is eliminated so that the common energy value is replaced by two different ones, one higher and one lower than it. The  $H_2$  molecule, *e.g.*, consists of two nuclei (*a*) and (*b*) and two electrons (1) and (2). Electron (1) may be with nucleus (*a*) and electron (2) with nucleus (*b*); the system may have an energy *E*. The electrons (1) and (2) are indistinguishable, so electron (1) may also be with nucleus (*b*) and electron (2) with nucleus (*a*) and the energy of the system will have the same value *E*. Wave-mechanics says then that the actual energy value is not *E*, but there are two possibilities one value lower than *E*, another higher.<sup>7</sup> The lower one is even lower than the sum of the energies of the two separated atoms and leads therefore to chemical union and the liberation of this energy difference appears as heat of formation. The higher one leads to an elastic collision of the two atoms. The non-elastic impact and chemical union occurs, when the two electrons possess anti-parallel spin vectors, the electrons then going into the same quantum group of the molecule, and the elastic impact occurs in the case of parallel spin vectors. The wave-function of the  $H_2$  molecule, corresponding to the lower energy value, shows a finite probability for the electron of the one atom to be also with the other one and we may interpret this as the analytical representation of Lewis' process of sharing. If on the other hand the same calculation is applied to two unexcited helium atoms, each possessing already two equivalent electrons in the same quantum group, *i.e.*, with counter-balanced spin, no splitting of the energy value occurs and chemical union is not possible. It can take place only between excited He atoms in which the closed group of electrons is fissured and indeed such  $He_2$  molecules formed by excited atoms are spectroscopically known to exist in the electric discharge tube. A closed quantum group always renders the atom chemically inert.

The wave-mechanical calculation, the results of which have been just described, was first given by Heitler and London. Since

<sup>7</sup> W. Heitler and F. London, *Zs. f. Phys.*, 1927, 44, 455.



the system of two hydrogen atoms contains four particles, the Schrödinger equation cannot be solved directly but has to be approximated. In the following table we compare the observed constants of the  $H_2$  molecule with the results of the calculation of Hylleraas,<sup>8</sup> who continued the wave-mechanical treatment with higher approximations.

	Energy of dissociation in electron volts (1 e. $v=23$ k cal/mol.)	Internuclear distance in Angstrom units ( $10^{-8}$ cm.)	Moment of inertia in g. cm. <sup>2</sup>
Calculated	4.37	0.72	$4.28 \times 10^{-14}$
Observed	4.4	0.76	$4.72 \times 10^{-14}$

Wave-mechanically the molecule is described by its wave-function. The method of Heitler and London consists in constructing the wave-function of the molecule by combining those of the separated atoms. Another way of doing it is to consider only the wave-functions of the valency electrons of the separated atoms and to build up that of the molecule from them. This method was successfully used by Slater and Pauling<sup>9</sup> in explaining the valency angle in such molecules as  $H_2O$ , in which the valency electrons of the central atom are  $p$ -electrons.

Both these methods commence with the system of the separated atoms, which are thought to approach each other, their mutual interaction gradually increasing. There is, however, a third method, developed mainly by Herzberg, Hund, Lennard-Jones and Mulliken,<sup>10</sup> which considers the already combined atoms, *i.e.*, the completed molecule from the very beginning and which is called the method of molecular orbitals, an orbital being a quantum group of the molecule. The nuclei are thought to be fixed at a particular internuclear distance together with their cores of inner electrons which do not take part in the chemical linkage, the so-called "atomic orbitals". The outside electrons are added one by one to this skeleton, filling up the "molecular orbitals". Thus the wave-function of the molecule as a whole is constructed

by combining those of the single electrons, which from the very beginning are under the influence of both the nuclei.

Ultimately of course all these methods are bound to merge and it should not matter, if the wave-function of the molecule is approximated by separated atoms, decreasing their distance, or by electrons brought in the field of the two nuclei which have already the proper internuclear distance prevailing in the molecule. We are, however, not able to calculate sufficiently high approximations and therefore the results may vary, each method giving a different approximation of the same true wave-function of the molecule. In general we may say that the results of the first two methods, which commence with the separated system, describe better the conditions in the molecule at larger distances of the nuclei, whereas the third method naturally gives better results at lesser distances. Since, however, actual calculation is possible only in the simplest cases, and has to be replaced by generalisation in heavier, poly-electronic molecules, there remains a certain discretion of interpretation, which leads to different theories of valency.

It is obvious that the first and second methods of mathematical treatment lead to an electron pair bond theory of valency; in poly-atomic molecules these pairs of electrons which represent the chemical bond are localised between two nuclei. Only the third method presents different ways of interpretation as to a theory of valency. Chemical linkage is always due to a degeneracy, as described above. In those simple cases, in which actual wave-mechanical calculation is possible, there exist, however, two possibilities. The linkage, *i.e.*, of  $H_2$  may be ascribed either to the degeneracy produced by the equality of the two electrons or to that produced by the equality of the protons. Hund has shown that the latter one is preserved to a certain extent, even when the nuclear fields are not exactly but almost equal. Assuming that chemical linkage is always due to the strict or approximated equality of the nuclear fields, we derive a theory of valency in which already the single electron produces a bonding effect; if, however, the generalisation is done on the lines, that the equality of the electrons produces the degeneracy necessary for chemical linkage, we are led to a pair bond theory of valency. In other words, when constructing the wave-function of the molecule by introducing the electrons one by one into the

<sup>8</sup> E. A. Hylleraas, *Zs. f. Phys.*, 1931, 71, 739.

<sup>9</sup> J. C. Slater, *Phys. Rev.*, 1931, 37, 481; 38, 328, 1109; 1932, 41, 255; L. Pauling, *Phys. Rev.*, 1932, 40, 891; *J. A. C. S.*, 1932, 53, 1367, 3225.

<sup>10</sup> G. Herzberg: *Zs. f. Phys.*, 1929, 57, 601. F. Hund: *Zs. f. Phys.* 1931, 73, 1, 565; 74, 1, 429. J. E. Lennard-Jones: *Trans. Farad. Soc.*, 1929, 25, 668; 1934, 30, 70; R. S. Mulliken: *Phys. Rev.*, 1932, 40, 55; 41, 49, 759; 1933, 43, 279 a.o.



combined field of the two nuclei, we may either consider them as independent, more or less neglecting their mutual interaction, or we may assume that such an approximation is insufficient for a theory of valency and that the mutual interaction is just the essential point for it. There is no doubt that both effects are always present but which of them prevails normally cannot be decided by the mathematical treatment itself. In any case, whether the wave-mechanical method of molecular orbitals is interpreted as a single electron bond theory of valency or as an electron pair bond theory, a postulate is introduced and the decision has therefore to be arrived at by comparing the results of both views with empirical facts.

It is already possible to calculate some of the important features of a molecule by treating the electrons as independent. The method of molecular orbitals owes its success just to this, that the term system and the electronic configuration of the completed molecule can be derived already in an approximation in which the interaction of the molecules has not to be considered. The results, obtained in this way, have led some authors to believe that the question of chemical linkage may also be reduced to the bonding effect of a single electron in degenerated nuclear fields. This view is supported by the existence of the molecule ion  $H_2^+$  which contradicts any pair bond theory of valency, because only one electron is left and here the linkage is certainly due to the strict degeneracy of the nuclear fields. In poly-atomic molecules the independent electrons are "non-localised" and do not belong to any particular nucleus, and this gives an opportunity to translate the Octet Theory of Chemistry into wave-mechanical language. It requires, however, the additional hypothesis, that the vastly different fields of say,  $C^{2+}$  and  $O^{4+}$  in CO or  $Ca^{2+}$  and  $F^{5+}$  in CaF etc. are almost degenerated.

As soon, however, as the nuclei are not protons, just those molecules exist which possess a bonding pair of electrons, as  $Li_2$ ,  $LiH$ ,  $BeH$ ,  $(BeH)^+$ , but those with one electron only cannot be found even spectroscopically like  $(Li_2)^+$ ,  $(LiH)^+$  or  $(BeH)^{++}$ . Furthermore some molecules possess excited electronic terms in which their energy of dissociation is considerably increased as compared with that for the ground level, and it was found that this phenomenon occurs just with molecules possessing free

valencies.<sup>11</sup> It was possible to give a simple explanation from the pair bond view, whereas the other viewpoint is at a loss to explain this phenomenon in a simple way. Similar results obtain with regard to the linkage of atoms, possessing a helium-like configuration of two s-electrons, like the atoms of the second group or C in  $CO_2$ .<sup>12</sup> The  $s^2$  group acts repulsively according to spectroscopical evidence and wave-mechanical calculation and these bonds can be understood easily in a pair bond theory as arising from excited atoms, whereas the single electron bond interpretation of the theory should expect a different term as the ground level of these molecules. As regards the point of view of chemistry, it has been shown recently that there exists no experiment which proves the Octet Theory and that experimental evidence favours rather a uniform pair bond theory.<sup>13</sup>

The question as to whether non-localised wave-functions, i.e., independent electrons, or localised functions, i.e., electron pairs with strong mutual interaction, describe the molecule better, can be decided by the incapability of the non-localised functions, to describe the process of dissociation. The view of the single electron bond theory leads automatically to an incorrect statement as to the products of this process. If in  $H_2$  the two electrons are treated as independent, the products of dissociation are 50%  $H+H$  and 50%  $H^+ + H^-$ , which of course is far from the truth. The same obtains in poly-atomic molecules. This takes place, because the non-localised wave-function of the molecule contains also the ionic terms of  $H^+$  and  $H^-$  in too high a percentage, and this is a direct expression of the independence of the electrons. If the two electrons have no interaction other than a screening effect, then indeed the probability of either electron, to go with one or the other of the nuclei, is always 50% and it does not matter, if the other electron is already in the vicinity of this particular nucleus or not. If there exists, however, a strong mutual influence between the two electrons, the choice of the second electron, to go with a particular

<sup>11</sup> H. Lessheim and R. Samuel, *Zs. f. Phys.*, 1933, **84**, 637; **88**, 276.

<sup>12</sup> H. Lessheim and R. Samuel, *Proc. Phys. Soc.* (London), 1934, **46**, 523.

<sup>13</sup> R. F. Hunter and R. Samuel, *J. C. S.*, 1934, 1180; *Chem. and Ind.*, 1935, **54**, 31. *Rec. Trav. Chim.*, P. B., 1935, **54**, 114.



nucleus, will depend also on the choice of the first one and the probability of going to that particular nucleus, to which also the other electron belongs, will be much less than that of going to the nucleus which is still without an electron. The pair bond interpretation naturally pictures the molecule with a strong interaction inside the electron pairs and a weak interaction from pair to pair. Therefore neither strictly localised wave-functions, in which the interaction from pair to pair is missing and not containing the ionic terms at all, nor strictly non-localised functions without any interaction but with the full weight of the ionic terms are a correct description. Slater therefore could show that in the first wave-mechanical method about which we spoke above, a certain percentage of the ionic terms has to be introduced, or in the molecular orbital method, the excessive influence of the ionic terms has to be cancelled to come to satisfactory results with regard to dissociation. Thus the two views approach each other.<sup>14</sup> Sometimes it was believed that in poly-atomic molecules the localised wave-functions of the pair bond interpretation represent a poorer mathematical approximation. If it would be true that the approximation by non-localised functions is mathematically the better one, this would be indeed a serious argument against the pair bond view. The discussion concerned particularly the linkage between two atoms with one *s*-electron each and a central atom with two *p*-electrons, the linkage  $s-p^2-s$ , as in  $H_2O$  or  $CH_2$ . It could, however, be shown that those functions which form the somewhat poorer approximation have still the disadvantage that they yield wrong percentages of dissociation products. If a different procedure, which gives the correct products of dissociation, is followed, wave-functions are obtained which yield not only almost localised bonds but also an improvement of the approximation.<sup>15</sup>

<sup>14</sup> J. H. Van Vleck, *J. Chem. Phys.*, 1933, 1, 177, 219; 1934, 2, 20.

<sup>15</sup> H. Lessheim and R. Samuel, *Proc. Ind. Acad. Sci.*, 1935, 1, 623. *Nature*, 1935, (Febr. 9th), p. 230.

Thus we see that in the method of molecular orbitals two interpretations are possible and these represent just those two schools of thought, which existed already in pre-wave-mechanical theories of linkage. The interpretation by independent electrons is certainly sufficient for the description of the completed molecule, but as soon as any question connected with the transition to greater internuclear distances, *i.e.*, with the dissociation of the molecule arises, the interpretation by the electron pairs is superior both from the viewpoint of mathematics and experimental physics. The theory of valency is certainly concerned more with the process of dissociation or formation than with the description of the completed molecule. Combining all the different points of view, chemical evidence, band spectra, and the analytical representation of the molecule, it can be shown that the pair bond interpretation results in a self-contained theory in which all the difficulties of the other view disappear.<sup>17</sup> To our mind, therefore, the balance of probability rests with the pair bond theory and we have to picture the linkage in a normal molecule like  $SF_6$  as produced by six bonds each of them almost completely localised between the S and one of the F nuclei. There is no difference at all between these six linkages. The real field of application of the non-localised wave-functions and many a beautiful explanation of complicated chemical questions by them is given in the aromatic substances, where indeed the electrons of the benzene ring have to be considered as non-localised, *i.e.*, as belonging equally to all the carbon atoms. The degeneracy of the nuclear fields on the other hand answers mainly for those effects, which are connected with the polarity of the molecule, as the transition from covalent to electrovalent linkage or the inductive effect, used in modern organic chemistry. With these questions we cannot deal here. If, however, we confine ourselves to normal molecules, it appears as if a uniform pair bond theory will answer best the requirements of chemistry and physics.