

Abegg, Lewis, Langmuir, and the Octet Rule

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Contrary to the virtually universal opinion of introductory chemistry textbook authors, the octet rule was not invented by G. N. Lewis.¹ Without a doubt Lewis' concept of the shared electron pair bond played a key role in giving the octet rule its present form. However, Lewis himself remained curiously ambivalent about the importance of the rule, and the recognition of the significance of the number eight in valence relationships actually has a long history that extends back to the last quarter of the 19th century and the work of Mendeleev on the periodic table. Though at first glance such ancient origins might seem surprising, a moment's reflection on the necessary relation between valence and chemical periodicity makes this connection quite reasonable.

Early History of the Octet Rule

In 1871 Mendeleev (Fig. 1) published an extended review on the chemical and physical periodicity of the elements, which, in revised form, served as the core for the chapter on the periodic table in his classic textbook "The Principles of Chemistry" (1, 2). Among the properties discussed was, quite naturally, the topic of valence, and Mendeleev enunciated two rules relating periodicity and valence in which the number eight played a key role. The first of these limited the maximum valence displayed by a given element to one of eight possible valence types, RX_n (where R is the element in question and X a univalent ligand or test element) and, by implication, asserted that the maximum possible valence of any element in the periodic table could never exceed eight. Although Mendeleev was unable to illustrate the entire series of maximum valence types using a single univalent test element

¹ A review of 20 recently published introductory college-level chemistry texts showed that all of them explicitly or implicitly attributed the octet rule to Lewis.

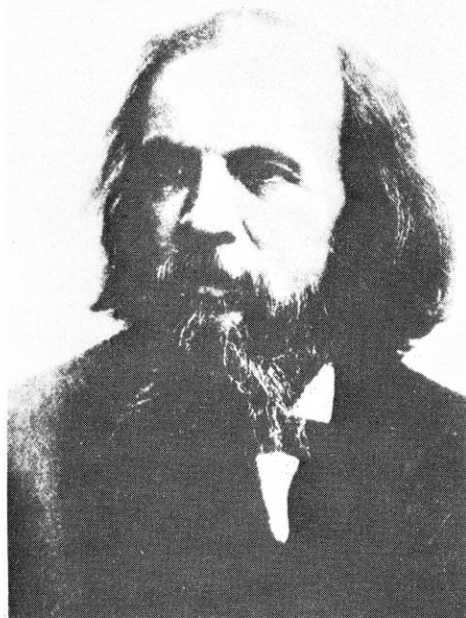
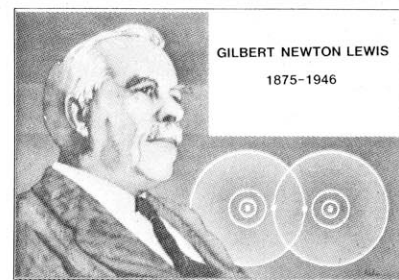


Figure 1. D. Mendeleev.



(Table 1, row 1), he was able to illustrate the entire series using bivalent oxygen as the test element (Table 1, rows 2 and 3), where the examples for the RO_4 ($\cong RX_8$) class were taken from the triad of elements headed by iron, cobalt, and nickel and belonging to what is today called group VIII.

Mendeleev's second rule established a relationship between an element's maximum valence as measured relative to hydrogen and its maximum valence as measured relative to oxygen, asserting that their sum was never greater than eight and that, indeed, for elements in groups IV through VII (so-called higher types) it was in fact equal to eight. In other words, if a given element of higher type had a hydride of the form RH_n , its highest oxide necessarily had a formula of the form R_2O_{8-n} (Table 2).

Though receiving wide circulation through the medium of his extremely popular textbook, Mendeleev's initial obser-

Table 1. Mendeleev's First Rule

Univalent:	RX	RX ₂	RX ₃	RX ₄	RX ₅	RX ₆	RX ₇	RX ₈
Divalent:	R ₂ O	RO	R ₂ O ₃	RO ₂	R ₂ O ₅	RO ₃	R ₂ O ₇	RO ₄
Example:	K ₂ O	CaO	B ₂ O ₃	SiO ₂	P ₂ O ₅	SO ₃	Cl ₂ O ₇	OsO ₄

Table 2. Mendeleev's Second Rule

Hydride RH_n	Oxide R_2O_{8-n}
CH ₄	C ₂ O ₄ \cong CO ₂
NH ₃	N ₂ O ₅
SH ₂	S ₂ O ₆ \cong SO ₃
ClH	Cl ₂ O ₇
none	OsO ₄



Figure 2. R. Abegg.

vations on the role of the number eight in valence relations did not receive further elaboration until 1902, when they came to the attention of a 33-year-old professor of chemistry at the University of Breslau named Richard Abegg (Fig. 2). Abegg was originally trained as an organic chemist, taking a PhD under A. W. Hofmann in 1891 for a dissertation on amidochrysene. However, shortly after graduation Abegg shifted to the field of physical inorganic chemistry, doing post-doctoral work under Ostwald, Arrhenius, and Nernst, and eventually becoming Nernst's assistant at Göttingen. There he specialized in the electrochemistry of both simple and complex ions and was naturally led to a study of those factors determining the ability of an ion to form complexes and to speculation on the possible electrical origin of chemical valence (3).

In 1899 Abegg published a joint paper with Guido Bodländer attempting to correlate the stability of complexes with the "electroaffinities" of the constituent ions and these, in turn, with the ions' redox potentials (4). In 1902, in the course of a lecture at the University of Christiania, Abegg extended this work to the more general problem of the origin and periodicity of chemical valence and postulated his at-one-time-famous rule of normal and contravalence (5).

Revising the earlier electrochemical dualism of Berzelius and Davy, Abegg postulated that all elements were capable of exhibiting both a maximum electropositive valence and a maximum electronegative valence and that the sum of the two was always equal to eight (Table 3). The maximum positive valence of an element was identical to its group number N and its negative valence to $8 - N$. Whichever of the two valences was less than 4 corresponded to the element's normal valence (as exhibited in the vast majority of its stable compounds), whereas the complementary valence corresponded to its contravalence (as exhibited in less stable, rarer combinations). For elements in group IV (e.g., silicon and carbon) there was no natural preference for either of the two valence types and such elements tended to be amphoteric (a term introduced by Abegg). Abegg was, of course, aware that in practice many elements failed to exhibit the full range of their potential valences. Indeed, he noted that elements in groups I-III never exhibited their contravalences (thus in effect restricting his law to Mendeleev's higher types) and that in the other groups the tendency to do so generally increased as one moved down the group (compare, for example, fluorine, with just HF, to iodine, with both HI and IF₇).²

Mendeleev's original formulation of his hydrogen-oxygen rule was without electrochemical implications. His maximum valence was strictly a stoichiometric valence derived from the classic relation

$$\text{valence} = \frac{\text{atomic weight}}{\text{equivalent weight}} \quad (1)$$

Within the context of Abegg's theory, however, hydrogen became a test element for establishing an element's negative valence, oxygen a test element for establishing an element's positive valence, and their sum a reflection of Abegg's rule of normal and contravalences.

Abegg's views were elaborated in greater detail in a long (50 pages) paper published in 1904 (6). Most of this paper dealt

² In a short note published in 1902 (*Chem. News*, 64 (August 8, 1902)) the British chemist Geoffrey Martin suggested that the recent discovery of various intermetallic compounds by Kurnakov had finally revealed species in which the elements of groups I-III displayed their maximum valence (or contravalence in Abegg's terminology). Quite independently of Abegg, Martin generalized Mendeleev's earlier observations with the equation and statement: "Hence, $V_1 + V_2 = 8$, i.e., the sum of the highest and lowest degrees of valence with which an element acts towards other elements is a constant whose value is 8." Martin also noted that "An element tends to act toward radicals of like electrical sign with its highest valence, but toward radicals of opposite electrical sign with its lowest valence," thus placing an electrochemical interpretation on Mendeleev's rule equivalent to that of Abegg.

with various experimental criteria for establishing the relative polarity of an element's valence in a given compound. However, near the end of the paper Abegg ventured some speculations on the possible origins of polar valence. Though not adverse to the concept of both positive and negative electrons, he tended to favor a unitary theory in which positive charge was due to electron deficiency, and within this context gave a simple and prophetic interpretation of his rule (6):

The sum 8 of our normal and contravalences possesses therefore simple significance as the number which for all atoms represents the points of attack of electrons, and the group-number or positive valence indicates how many of these 8 points of attack must hold electrons in order to make the element electrically neutral.

Unlike most other chemists and physicists encountered in the history of the octet rule, Abegg is now virtually forgotten, though there can be no doubt that his rule formed a key step in the development of both the octet rule and the electronic theory of valence (7). The rule was explicitly referred to by all of the pioneers in the field who followed Abegg and, indeed, served as the "chemical law of valence" against which they tested their own theories of valence and atomic structure. The most likely reason for Abegg's eclipse was his premature death at age 42 in a ballooning accident, and it is tempting to speculate on how the history of the electronic theory of valence might differ had he lived, especially if he had been able to modify his initial commitment to an extreme dualism (which, given his training as an organic chemist, is not improbable). Interestingly, at the time of his death, Abegg was, in true Germanic fashion, in the process of editing an enormous multivolume "Handbuch der Anorganische Chemie" in which he was attempting to use his rule as a unifying theme to organize the descriptive chemistry of the elements (8).

Abegg's rule established an explicit connection between the numerical limits of valence and the electronic theory of matter, then in its infancy. However, it did not attempt to provide an explicit model of atomic structure, contenting itself instead

Table 3. Abegg's Rule of Normal and Contravalence (6)

	Gruppe:						
	1	2	3	4	5	6	7
Normalvalenzen	+1	+2	+3	±4	-3	-2	-1
Kontravalenzen	(-7)	(-6)	(-5)	±4	+5	+6	+7



Figure 3. J. J. Thomson.

with the vague concept of "points of attack." This deficiency was made up in large part by the work of the English physicist J. J. Thomson (Fig. 3). Ever since his discovery of the electron in 1897 (which for some peculiarly British reason he persisted in calling a negative corpuscle—a term more appropriate to Boyle and the 17th century than to 20th century physics), Thomson had doggedly pursued a workable electronic model of the atom. By 1907 his search had given birth to the famous plum-pudding model and to at least four valuable concepts which, though no longer used in the explicit forms given them by Thomson, are nevertheless an implicit part of all currently accepted electronic theories of valence: 1) the concept of an electronic shell structure for the atom; 2) the concept that chemical valence is largely a function of only the outermost electronic shell; 3) the concept that chemical periodicity implies a periodic repetition of the outer electronic shell structure; and 4) the concept that the stability of the rare gases is connected with shell completion and that the valence of other atoms can be correlated with an attempt to attain similar closed-shell structures via electron transfer (9).

In a manner unusual for a physicist, Thomson was quick to point out the chemical implications of his model, explicitly connecting it with Abegg's rule and in the process enshrining the number eight as the central numerical deity of chemical valence (10). Summarizing his views in 1914, he also added what would, with the gift of historical hindsight, prove to be an interesting caveat (11).

We regard the negatively electrified corpuscles in a atom as arranged in a series of layers, those in the inner layers we suppose are so firmly fixed that they do not adjust themselves so as to cause the atom to attract other atoms in its neighbourhood. There may, however, be a ring of corpuscles near the surface of the atom which are mobile and which have to be fixed if the atom is to be saturated. We suppose, moreover, that the number of corpuscles of this kind may be anything from 0 to 8, but that when the number reaches 8 the ring is so stable that the corpuscles are no longer mobile and the atom is, so to speak, self-saturated. The number of these mobile corpuscles in an atom of an element is equal to the number of the group in which the element is placed on Mendeleev's arrangement.

... Thus we see that an atom may exert an electro-positive valence equal to the number of mobile corpuscles in the atom, or an electronegative valence equal to the difference between eight and this number. Each atom can, in fact, exert either an electro-positive or electronegative valency, and the sum of these two valencies is equal to eight. In this respect the theory agrees with Abegg's theory of positive and negative valency. ... It is possible that the number of corpuscles which form a rigid ring may depend to some extent on the number in the inner rings, i.e. on the atomic weights of the elements, and that for elements with atomic weights greater than forty (i.e., Ar) this number may not be eight. If this should prove to be the case, the sum of the positive and negative valencies for such elements would not be equal to eight.

Kossel and Lewis

The year 1916 saw the publication of two extremely important papers on the electronic theory of valence. The first, published in the March issue of the *Annalen der Physik*, was by a 28-year-old physicist at the University of Munich named Walter Kossel (Fig. 4). Kossel's paper, or rather his monograph (133 pages!), was an explicit attempt to develop and extend Abegg's valence views for relatively polar inorganic compounds using the electron transfer model briefly hinted at by Abegg near the close of his 1904 paper (indeed Abegg is explicitly cited by Kossel no less than 20 times (12)). In keeping with this goal, Kossel was less interested in an explicit atomic



Figure 4. W. Kossel.

model (though he was fully aware of the Bohr-Rutherford model) than in accurate experimental counts of the total number of electrons per atom, data not available to Thomson in 1907. These he obtained from the work of the physicist A. van den Broek and used to construct his now famous plot of maximum positive and negative oxidation states for the first 57 elements (Fig. 5). This was a plot of the total electron count per atom or ion versus its atomic number, with the values for the neutral elements themselves appearing along the diagonal, those for the elements in their maximum positive oxidation states appearing as extensions below the diagonal, and those for their maximum negative oxidation states as extensions above the diagonal.

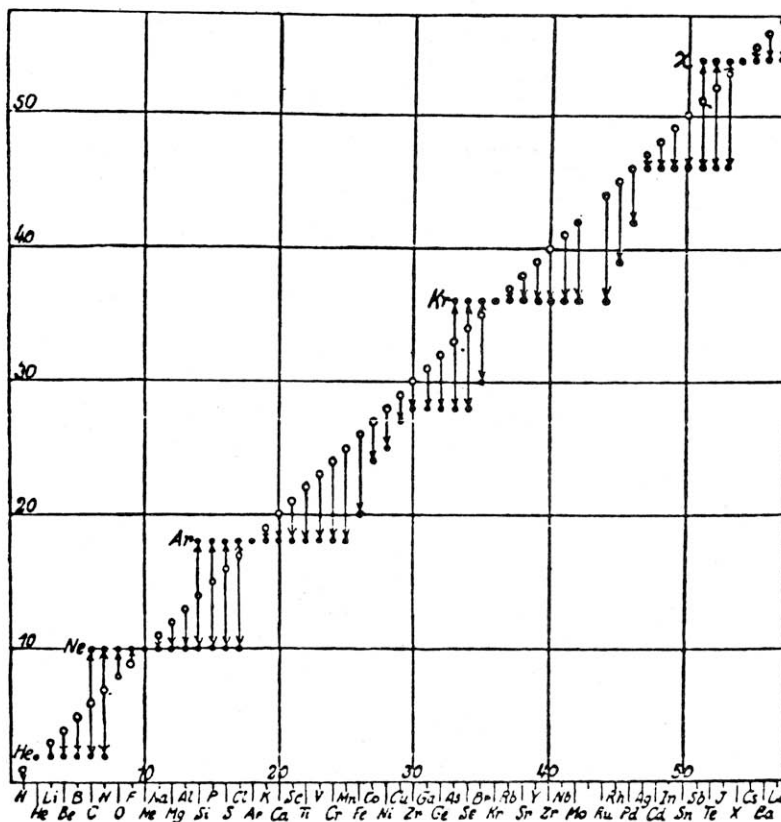


Figure 5. Kossel's plot of maximum positive and negative oxidation states (12).

This plot allowed Kossel to assess in one stroke the validity of both Abegg's rule and Thomson's suggestion that electron transfer was associated with attempts to attain a closed-shell, rare gas structure. As can be seen from the figure, both Thomson's postulate and Abegg's rule held fairly well for the *s*- and *p*-block elements (using modern terminology) as indicated by the clumping of the oxidation states about the rare gas counts, and the symmetrical disposition of oxidation states on either side of the diagonal for elements in groups IV through VII. However, problems were readily apparent for the *d*-block elements. Although the elements up to group VIII appeared to attain a maximum positive oxidation state corresponding to a rare gas electron count, in the case of groups IVA–VIII (e.g., Ti–Mn) they failed, in violation of Abegg's rule, to attain the corresponding negative states of their group B analogs³ (e.g., Si–Cl), a fact which Kossel felt to be of a great significance in differentiating the two classes of elements.

Beginning with group VIII there was an erratic variation in the maximum oxidation state, culminating in a second asymmetrical clumping of positive oxidation states about the counts for the last members of the group VIII triads (e.g., Ni and Pd). Today we associate these clumpings with the attainment of a pseudo-rare gas ($n - 1$)*d*¹⁰ configuration. However, since Kossel was using total electron counts, rather than configurations, the 10-valence electron count became associated with Ni and Pd instead. This led Kossel to postulate that these elements represented a kind of imperfect or weakened analog of the rare gases as, like the rare gases themselves, they bridged the gap between one cycle of repeating maximum oxidation states (e.g., K[I]–Mn[VIII]) and another (Cu[II]–Br[VII]). The striving for a maximum positive oxidation state corresponding to either a rare gas or pseudo-rare gas electron count was virtually universal. The striving for a maximum negative oxidation state corresponding to a rare gas electron count, however, fell off rapidly as one moved to the left of the rare gases and was nonexistent in the case of pseudo-rare gas counts, thus accounting for the asymmetry in the behavior of the group A *d*-block elements. In no case did the maximum oxidation state of an element exceed eight.

The rest of Kossel's paper was essentially an application of the resulting ionic bonding model to the problems of coordination chemistry, the autoionization of protonic solvents, and the relative acidity and basicity of oxides. Also included were some simple calculations using Coulomb's law and an attempt to visualize the resulting bonds in terms of Bohr's atomic model. When combined with Fajans' later work on ionic po-



Figure 6. G. N. Lewis.

larization, Kossel's treatment of coordination compounds proved quite useful and was popular among European chemists up until the Second World War, though it never attained a similar popularity among American and British chemists (13).

The second 1916 paper, published one month after Kossel's in the *Journal of the American Chemical Society*, was by a 40-year-old chemist at the University of Berkeley named G. N. Lewis (Fig. 6) and, in contrast to Kossel's epic, was quite terse (22 pages) (14). Lewis had actually begun speculating on the electronic theory of valence as early as 1902 (about the year of Abegg's lecture at Christiania) and had independently arrived at a dualistic electron-transfer model similar to Abegg's. However, like Thomson, Lewis had also proceeded to develop a specific static model of the atom, which he called the cubical atom, and had thus apparently also independently arrived at the concept of periodically repeating electronic shells and the concept of the relative stability of rare gas structures (Fig. 7).

Fortunately Lewis became increasingly skeptical about the ability of the electron-transfer model to account for the relatively nonpolar compounds of organic chemistry and by 1913 was willing to publish a paper stating his belief that two radically distinct kinds of bonds were required in chemistry, the polar electron-transfer bond of typical ionic salts and some kind of nonpolar bond to represent the valence stroke of structural organic chemistry (15). Largely for this reason, apparently, Lewis was unwilling to publish his cubical atom model.

However, some time between 1913 and 1916 Lewis was able to resolve his dilemma by using the cubical atom to arrive at the concept of the shared electron pair bond (Fig. 8). The resulting "simple assumption that the chemical bond is at all times and in all molecules merely a pair of electrons jointly held by two atoms" not only provided the missing bond mechanism for the nonpolar valence stroke, but also allowed Lewis to resolve the dichotomy of two distinct bond types "so repugnant to that chemical instinct which leads so irresistibly to the belief that all types of chemical union are essentially one and the same," through his realization that the progressively uneven sharing of the electron pair between atoms of in-

³ According to IUPAC notation.

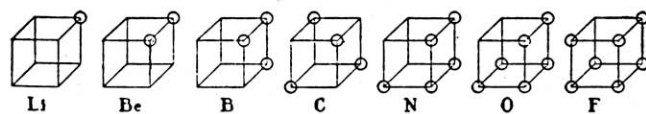


Figure 7. Lewis' cubical atom (14).

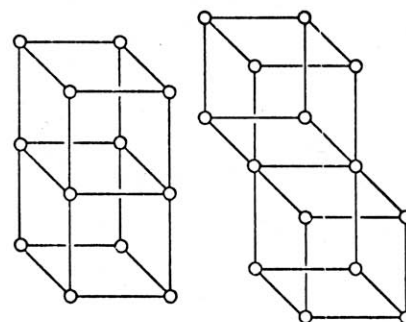


Figure 8. Single and double electron pair bonds viewed using the cubical atom (14).

creasingly different electronegativities could yield the ionic electron-transfer bond as a limiting case.

The idea that a chemical bond could result from the sharing as well as from the transfer of electrons was not unique with Lewis. The chemists A. L. Parson (16), W. C. Arsem (17) and H. Kauffmann (18), as well as the physicists J. Stark (19) and J. J. Thomson himself (11), had suggested similar ideas. The difficulty with all of these proposals, however, was that no reasonable restraints had been placed on either the total number of electrons per bond or on the number of atoms that could share a common electron. As a consequence the resulting models were too open-ended to be operationally useful. Lewis, by restricting both the number of electrons per bond and the number of atoms so bonded to two, provided the necessary restraints to give a workable theory. Robert Kohler, who has studied the origins of the shared electron pair bond, has suggested that the concept resulted from Lewis' attempts to accommodate the electron-sharing concepts of Parson and Thomson within the context of his own dualistic cubical atom model, and that the restraints so necessary to its success were a natural consequence of the geometrical restraints imposed by the cubical atom itself (20).

It is worth pointing out that Kossel had also recognized the necessity of a nonpolar bond for organic compounds as well as the concept of a progressive change in bond type. Early in his paper Kossel had noted that the electron-transfer model was unsuitable for nonpolar organic compounds and that an electron-sharing model similar to Stark's would probably be required. Near the end of the paper Kossel proposed some specific models using two-dimensional Bohr-Rutherford ring atoms in which all of the valence electrons (e.g., 10 in the case of N_2) were held on a common ring placed symmetrically between the two bonded atoms (Fig. 9). As the polarity of the bond increased, this ring was progressively displaced toward the more electronegative atom, and in the ionic limit became its sole property.

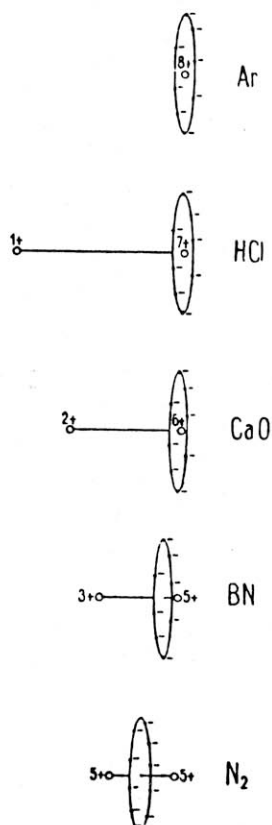


Figure 9. The progressive change from ionic to covalent bonding in terms of Kossel's ring atom model (12).

Like his predecessors, Lewis had recognized both the importance of the number eight in valence relationships (which he called the rule of eight) and its correlation with the attainment of rare gas structures. However, within the context of the ionic model, as formulated by Kossel, this correlation was only possible in the case of maximum oxidation states (e.g., P^{5+} and Cl^- in PCl_5). Attempts to construct ionic formulas for lower oxidation state compounds failed to give ions having either rare gas or pseudo-rare gas electron counts (e.g., P^{3+} in PCl_3). The importance of the shared electron pair bond for the octet rule was that it allowed the extension of the rule to these lower oxidation state species as well as to purely homonuclear species such as Cl_2 and N_2 . In contrast, Kossel's formulation of homonuclear species precluded their obeying the octet rule (e.g., N_2) and, indeed, Kossel went so far as to suggest that the rule was limited only to polar compounds. Though Lewis was well aware of the implications of the electron pair bond for the octet rule, their full exploitation was to be largely the work of Irving Langmuir.

Irving Langmuir

Irving Langmuir (Fig. 10) was a 38-year-old research chemist at General Electric when he first began to write on the subject of Lewis' electron pair bond in 1919 (21). In the course of three short years he managed, in addition to numerous popular lectures, to publish more than 12 articles, extending, refining, and popularizing Lewis' work. Lewis had originally restricted his discussion of the cubical atom and chemical bonding to the *s*- and *p*-block elements, feeling that the ambiguities then present in the chemistry and electronic structures of the *d*-block elements made attempts to extend the model too speculative. Langmuir, in his first and most ambitious paper (66 pages), published in the June 1919 issue of the *Journal of the American Chemical Society*, attempted to remedy this defect (22). Through the use of 11 basic postulates covering the arrangement of the electrons in isolated atoms and their preferential sharing in compounds, Langmuir undertook to deduce the entire structure of theoretical and descriptive chemistry. The full extent of his ambition is revealed in a popular lecture published in 1921 (23). Langmuir wrote



Figure 10. I. Langmuir.

These things mark the beginning, I believe, of a new chemistry, a deductive chemistry, one in which we can reason out chemical relationships without falling back on chemical intuition. . . . I think that within a few years we will be able to deduce 90 percent of everything that is in every textbook on chemistry, deduce it as you need it, from simple ordinary principles, knowing definite facts in regard to the structure of the atom.

Despite his enthusiasm, Langmuir's extension of the static atom (Fig. 11) proved to be short-lived. By 1921 Bohr, on the basis of spectral evidence (24) and Bury, on the basis of chemical evidence (25), had shown that Langmuir's postulated shell sequence of 2, 8, 8, 18, 18, and 32 electrons was wrong as was his postulate that a new electron shell could be started only if the preceding shells were completely filled. In addition, most theoretical physicists, who by this time were firmly committed to a dynamic atom model, were less than impressed by Langmuir's extended static model. The German physicist Sommerfeld characterized Langmuir's 11-postulate deductive chemistry as "somewhat cabalistic" (26), and the English physicist E. N. Andrade was even more blunt (26),

It is scarcely necessary to insist on the artificiality of this picture The electrons in Langmuir's atom have, in fact, so few of the known properties of electrons that it is not immediately clear why they are called electrons at all.

Thus, in actual practice, Langmuir's development of Lewis' model was restricted to the same atoms as Lewis had originally discussed. Nevertheless, a number of valuable concepts did evolve out of Langmuir's refinements, including the electroneutrality principle and the isoelectronic (isosteric) principle (27), the last of which has been characterized by Kober, in a recent article, as the "last important nonquantum mechanical bonding principle" (28).

Even more important to the present discussion was Langmuir's use of his postulates to deduce a general mathematical expression for the "octet" rule (a term introduced by Lang-

Table 4. Examples of Langmuir's Octet Equation

Species	Σe	n	H	$\nu = \frac{1}{2}(8n - \Sigma e) + H$	Structure
NH ₃	8	1	3	3	
CO	10	2	0	3	
H ₂ CO	12	2	2	4	
NaCl	8	1	0	0	Na ⁺ [Cl ⁻]
CS ₂	16	3	0	4	

muir to replace Lewis' more cumbersome "rule of eight")

$$\Sigma e = \Sigma s - 2B \quad (2)$$

where Σe is the total sum of available valence electrons in a species, Σs is the sum of the valence electrons required for complete shell formation for each of the heavier atoms (i.e., excluding hydrogen), and B is the number of covalent bonds between the heavier atoms. Restriction of the heavy atoms to those lying below Ar converts eqn. (2) into the major equation of Langmuir's "octet theory"

$$\Sigma e = 8n - 2B \quad (3)$$

when n is the number of heavy atoms requiring octet completion. Addition of the covalent bonds, H , to any protons present gives the final total number of covalent bonds in a species

$$\nu_{\text{cov}} = H + B = H + \frac{1}{2}(8n - \Sigma e) \quad (4)$$

Some example applications of eqn. (4) are shown in Table 4

He	Li	Be	B	C	N	O	F				
Ne	Na	Mg	Al	Si	P	S	Cl				
Ar	K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	
Ni/β	Cu	Zn	Ga	Ge	As	Se	Br				
Kr	Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	
Pd/β	Ag	Cd	In	Sn	Sb	Te	I				
Xe	Cs	Ba	La	Ce	Pr	Nd		Sa	Eu	Gd	
		Cells-32 Electrons-1	Cells-32 Electrons-2	Cells-32 Electrons-3	Cells-32 Electrons-4	Cells-32 Electrons-5	Cells-32 Electrons-6	Cells-32 Electrons-7	Cells-32 Electrons-8	Cells-32 Electrons-9	Cells-32 Electrons-10
		53	54	55	56	57	58	59	60	61	62

CHART I.—Illustrating the Lewis-Langmuir Theory of Atomic Structure.

(Facing page 470.)

and some examples of Langmuir's original structures in Figure 12.

In 1921, in what was essentially his last major paper on the octet theory, Langmuir gave a comprehensive survey of the limitations of eqn. (4) (29). By this time he had succeeded in reducing his original 11 postulates to three, the third of which represented the first explicit statement of the electroneutrality principle

- 1) The electrons in atoms tend to surround the nucleus in successive layers containing 2, 8, 8, 18, 18, and 32 electrons respectively.
- 2) Two atoms may be coupled together by one or more duplets held in common by the completed sheaths of the atoms.
- 3) The residual charge on each atom or group of atoms tends to a minimum.

Shell completion was desirable because it resulted in a spherically symmetrical electron distribution which tended to minimize external electrical fields about the atom. Minimization of the residual charges was also desirable because it minimized the coulombic energy required to maintain charge separation on adjacent atoms.

Langmuir pointed out that the requirements of postulates 1 and 3 were frequently in conflict and used this conflict to rationalize the limitations of his octet equation. His resulting classification of chemical compounds is shown in Table 5. Species obeying postulate 1 were termed "complete compounds" and tended to obey the octet rule either through the formation of ionic compounds (Class IA) or through the formation of "covalent" compounds (Class IB)—another term

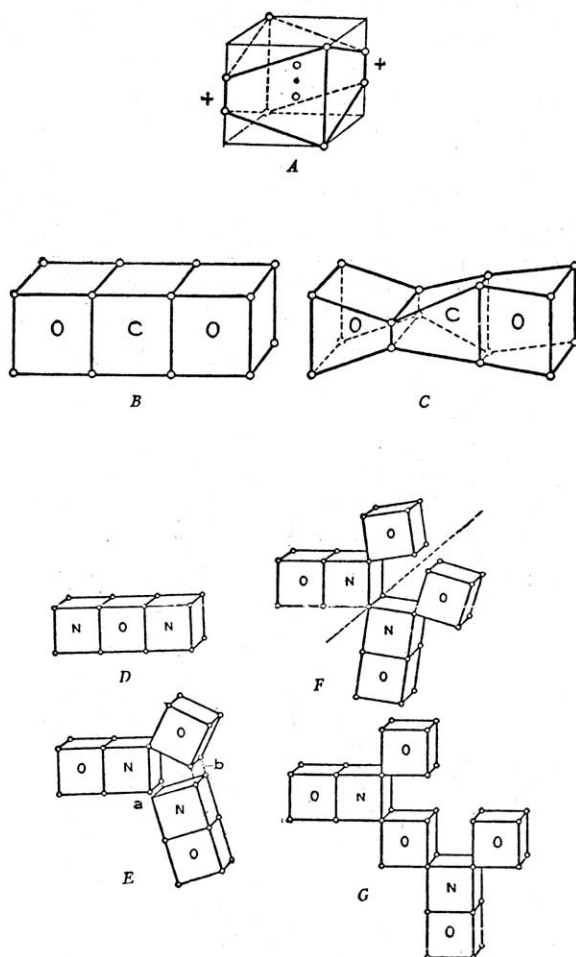


Figure 12. Some of the "cubical atom" molecular structures which Langmuir deduced using his octet equation (27).

introduced by Langmuir. In the latter case, Langmuir further distinguished between those species obeying the octet rule proper (i.e., $\sum s = 8n$) and those obeying an extended rare gas rule (i.e., $s = 18$), the former being restricted to combinations among elements corresponding to Mendeleev's higher types and the latter being the first explicit statement of the 18-electron rule much beloved of the organometallic chemist.

On the other hand, the "incomplete compounds" of Class II failed to obey postulate 1 and thus the octet rule. In Langmuir's opinion this was because shell completion in these species required, in violation of postulate 3, the buildup of prohibitively large net ionic charges (and here Langmuir was generous in allowing charges as high as 6+ and 4- in contrast to Pauling's later restriction of $2\pm$ or lower). Finally, Langmuir included a class of "exceptional cases" whose bonding apparently required a special kind of electron-sharing beyond that proposed in postulate 2.

The Debate

Lewis' opportunity to comment on Langmuir's work came in 1923 with the publication of his classic monograph, "Valence and Structure of Atoms and Molecules" (30). With the exception of a short retrospective essay written in 1933 (31), this was essentially Lewis' last major contribution to the electronic theory of valence and represented his matured views on the nature of the electron-pair bond.

Already in his 1916 paper, Lewis, faced with the inability of the cubical atom to represent a triple bond and the overwhelming evidence of organic chemistry that the valences of carbon were tetrahedrally directed, had speculated that the electrons of the cubical atom were drawn together to form a tetrahedron of electron pairs (Fig. 13). By 1923 Lewis had also abandoned his earlier defense of the static atom (32) and was willing to compromise with the dynamical atom models favored by the physicists, suggesting a model in which the electrons of the static atom corresponded to the average positions of electrons moving in small directional orbits. In keeping with these views, the cubical atom was presented only in an historical context in the 1923 monograph. Cubical

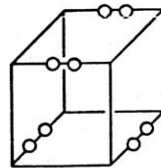


Figure 13. Lewis' tetrahedral atom with close-paired electrons (14).

Table 5. Langmuir's Classification of Compounds via the Octet Rule

I. Complete Compounds	
A) Compounds Without Covalence ($B = 0$)	
1) Electropositive main group elements ($N \leq 3$) plus electronegative elements ($N \geq 4$), e.g., NaCl, NB, Al_2O_3	
2) Early transition elements in their maximum oxidation state plus electronegative elements, e.g., $ScCl_3$, $TaBr_5$, CrF_6	
3) Certain halides of the electronegative elements in their higher oxidation states, e.g., PCl_5 , SF_6	
B) Covalent Compounds	
1) Among the electronegative elements, where $s = 8$, ($N \geq 4$), e.g., C, N, O, F, Si, P, Cl	
2) Among the transition elements, where $s = 18$, e.g., $Mo(CO)_6$, $Fe(CO)_5$ and $ni(CO)_4$	
II. Incomplete Compounds	
A) Intermetallics	
B) Salts of the later transition elements, e.g., $CrCl_3$, $CuCl_2$, ZnO	
III. Exceptional Cases	
A) Triply bonded species, e.g., N_2 , CO, CN^-	
B) Hydrogen-bonded species, e.g., HF_2^-	
C) Boranes	

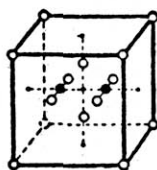


Figure 14. Langmuir's cubical structure for N_2 , CO , and CN^- (27).

models of molecules, such as those used by Langmuir (recall Fig. 12), were used nowhere in the book, extensive use being made instead of the less structurally explicit electron dot formulas, though in several places tetrahedral atom models were also used.

Langmuir's failure to abandon the cubical atom accounted for his classification of N_2 , CO , and CN^- as "exceptional cases." Lacking a triple bond, he was forced to postulate a special electronic structure for these species in which both kernels were simultaneously enclosed within a single cube of electrons, the remaining two electrons being shared as a normal duplet between the kernels (Fig. 14), a proposition which Lewis was to later characterize as "ad hoc" (30).

Though, as Kohler has suggested, the cubical atom probably played a central role in the initial inception of the electron pair bond, Lewis was quick to obtain an empirical base of support for the bond, the most impressive evidence being the observation that virtually all stable chemical species contained an even number of electrons. Indeed, the few "odd molecule" exceptions to this rule tended to be highly reactive and frequently underwent dimerization reactions to give products with even electron counts. Lewis also became increasingly convinced, in large part through the influence of A. L. Parson's magneton model of the atom, that magnetic interactions between electrons were responsible for the formation of electron pairs, a thesis discussed at length in a separate chapter of the 1923 monograph and in a review published the next year (59). These considerations led Lewis to postulate that the formation of electron pairs or the "rule of two," as he called it, was a much more important bonding principle than the rule of eight and account, in part, for his evaluation of Langmuir's octet equation. Lewis wrote (30)

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 overemphasizes what is after all but one feature of the new theory of valence. The rule of eight, in spite of its great importance, is less fundamental than the rule of two, which calls attention to the tendency of electrons to form pairs.

Langmuir, on the other hand, felt that Lewis' neglect of transition metal chemistry had misled him concerning the necessary instability of "odd molecules," and that their relative scarcity was due less to an inherent instability resulting from their unpaired electrons than to the relative abundance of closed-shell species, or in Langmuir's own words (29)

... the remarkable tendency, pointed out by Lewis, for most compounds to contain an even number of electrons is due merely to the relative abundance of complete compounds compared to incomplete ones. In other words, the even number of electrons in most compounds results from the tendency of Postulate 1 [i.e. closed-shell formation] rather than from any more general tendency for electrons to form pairs.

Thus Langmuir viewed the prevalence of even-electron-count species as a consequence of complete shell formation and as a reflection of the octet rule for elements lying below Ar, whereas Lewis viewed it (and the octet rule itself) as a consequence of the "rule of two."

Table 6. Lewis' Examples of Species Violating the Octet Rule

1) Odd Molecules:	NO , Cl_2O
2) Electron Deficient:	$NaCH_3$, BR_3 , BF_3 , SO_3 , HPO_3 , CO_3^{2-} , NO_3^- , possibly even $NaCl_{(g)}$
3) Electron Rich:	PCl_5 , SF_6 , UF_6 , SIF_6^{2-} , $PtCl_6^{2-}$ and $Mo(CN)_8^{4-}$

Lewis even included a chapter in his monograph dealing with "Exceptions to the Rule of Eight" in which he discussed the examples summarized in Table 6. In viewing this list one is struck by the fact that Langmuir's own discussion of this subject was in some ways more thorough. Although the odd molecules in Class 1 certainly violate the octet rule, their supposed scarcity and high reactivity could be used to support the stability of the octet just as well as the stability of the electron pair. On the other hand, all of Lewis' electron-deficient examples and most of his electron-rich examples could be (and were) accommodated by Langmuir's octet equation by using an ionic formulation. Finally, several of the electron-rich examples were not fair game (e.g., $PtCl_6^{2-}$, UF_6 , and $Mo(CN)_8^{4-}$) as Langmuir had never claimed that the octet rule was valid for elements above Ar.

Lewis' purpose was, of course, less to find examples which could not be fitted by Langmuir's equation than to find examples which could be fitted only by postulating structures (usually ionic) which were at variance with their known physico-chemical properties. Indeed, in looking at the positions taken by Langmuir and Lewis regarding the octet rule, one receives the impression that Langmuir, in his desire to emulate the rigor of physics, wished to be guided by the deductions of his postulates, whereas Lewis, more open to the experimental data base of chemistry, was more willing to be guided by the empirical evidence.

There is little doubt that the personal interaction between Lewis and Langmuir also tended to color the positions they took regarding the octet rule. Langmuir was a popular and dynamic lecturer and Wilder D. Bancroft has left us with an amusing picture of what it was like to be a member of one of Langmuir's audiences (33).

Langmuir is the most convincing lecturer I have ever heard. I have heard him talk to an audience of chemists when I knew they did not understand one-third of what he was saying; but thought they did. It's very easy to be swept off one's feet by Langmuir. You remember in Kipling's novel *Kim* that the water-jug was broken and Lurgan Sahib was trying to hypnotize Kim into seeing it whole again. Kim saved himself by saying the multiplication table ... [so] ... I have heard Langmuir lecture when I knew he was wrong, but I had to repeat to myself "He is wrong, I know he is wrong, he is wrong" or I should have believed like the others.

Naturally Lewis was initially quite pleased with the publicity given his theory as a result of Langmuir's lectures and papers. However, as time passed and more and more of the publicity accrued to Langmuir instead of Lewis, Lewis became increasingly upset, expressing his true feelings in a letter written to W. A. Noyes in 1926 (34).

Perhaps I am inclined to be too caustic in this matter, but I really do feel that while people were justified in being carried away a bit by Langmuir's personal charm and enthusiasm some years ago, to persist, especially as they do in England, in speaking of the Langmuir theory of valence is inexcusable.

Another of Langmuir's talents was the ability to coin catchy terminology, such as duplet, isostere, octet rule, and covalent bond, and it is not without note that the first two terms do not appear in the index of Lewis' monograph and that the terms octet theory and covalence appear only once. In fact Lewis recommended that the term covalent be dropped because Langmuir had "associated this term with an arithmetical equation by which he attempts to predict the existence or nonexistence of chemical compounds" (30). In private Lewis rather humorously expressed his resentment over the

manner in which Langmuir had ignored Lewis' own terminology (34),

... sometimes parents show a singular infelicity in naming their children, but on the whole they seem to enjoy having the privilege.

In public Lewis was, of course, less outspoken and in his 1923 monograph gave an evaluation of Langmuir's contributions which can only be viewed as a masterpiece of diplomatic understatement (30).

It is a cause of much satisfaction to me to find that in the course of this series of applications of the new theory, conducted with the greatest acumen, Dr. Langmuir has not been obliged to change the theory which I advanced. Here and there he has been tempted to regard certain rules or tendencies as more universal in their scope than I considered them in my paper, or than I now consider them, but these are questions we shall have a later opportunity to discuss. The theory has been designated in some quarters as the Lewis-Langmuir theory, which would imply some sort of collaboration. As a matter of fact Dr. Langmuir's work has been entirely independent, and such additions as he has made to what was stated or implied in my paper should be credited to him alone.

In the final analysis there can be little doubt that Langmuir played a key role in accelerating the acceptance of Lewis' electron-pair bond. Robert Kohler, who has studied the interaction between Lewis and Langmuir in some detail, has concluded that by 1920 the chemical community was still, by and large, content to use the ionic electron-transfer bond and that no crisis in chemical bonding was evident which might draw chemists to a model similar to Lewis' electron-pair bond. The widespread and rapid acceptance of the electron-pair bond was rather a consequence of Langmuir's dynamic personality and the unique manner in which he spanned both the industrial and university chemical communities. As Kohler has written (21)

What made the difference was, of course, Langmuir himself and his personal reputation. The new theory was well received because it was "Langmuir's theory" and not because of its intrinsic intellectual worth. Once received, of course, its worth become evident. What I wish to stress is that the advantages of the new theory were not immediately obvious and that had it not been "Langmuir's theory," the rediscovery and adoption of Lewis' theory might well have awaited a real crisis in the theory of bonding.

Summary, Conclusions, and Prospects

The major events in the development of the octet rule are summarized in Table 7, though obviously many additional contributions were made by lesser-known chemists which could not be touched upon for lack of space.⁴ These events allow one to draw at least three major conclusions about the origins and development of the octet rule:

- 1) Within the context of the dualistic or electrovalent model the octet rule essentially corresponded to a maximum valence rule only. While the striving for a rare gas structure determined both which elements could exhibit negative valences and the value of these valences, in the case of the positive valence it merely determined the maximum value, placing no necessary restrictions on the lower values.
- 2) The covalent model, by providing a means, via electron sharing, of rationalizing both the lower positive oxidation states and homonuclear species in terms of rare gas structures, substantially strengthened the identification of the octet rule with the formation of rare gas structures.
- 3) At no time, within the context of either the electrovalent or covalent model, was it claimed that the octet/rare gas structure relation was generally valid for elements beyond Ar.

⁴ Some minor contributions, though hardly by lesser-known chemists, include the work of Ramsay and Nernst. Ramsay was one of the first to apply Abegg's rule of eight, and Nernst expressed some suggestive views on the saturation of ions. For a discussion, see Stranges (7).

Table 7. Summary of Major Events in Development of the Octet Rule

<i>Mendeleev: 1871</i>
Eight as a maximum valence rule and the sum of the hydrogen and oxygen valences for higher types.
<i>Abegg: 1904</i>
Electrochemical interpretation of Mendeleev's rule of eight in terms of electron gain and loss.
<i>Thomson: 1904, 1907</i>
Concept of chemical periodicity in terms of recurring outer electron configurations. Rule of eight as striving for completion of stable rare gas shells.
<i>Kossel: 1916</i>
Extension of ionic model. Eight as a maximum valence rule for polar compounds only.
<i>Lewis: 1916</i>
Continuity of bond type and electron pair bonding mechanism for octet completion.
<i>Langmuir: 1919-1921</i>
Elaboration and popularization of the Lewis model. Mathematical formulation of the octet rule.

The debate over the validity of the octet rule did not, of course, end with Lewis and Langmuir. Between the 1920's and the 1940's a number of vigorous discussions took place in which not only the validity of the octet rule was questioned but the validity of the electron-pair bond itself (35-37), and aspects of this debate are still with us (38, 39). More recent quantum mechanical calculations, however, have tended to discount the importance of octet expansion and have reinstated the general validity of the octet rule for the *p*-block elements (40). In keeping with this conclusion, modern bonding theory has also provided a satisfactory resolution of the problems associated with both electron-deficient and electron-rich species and has done so in a manner which shows that neither Lewis nor Langmuir was totally correct in the positions they took. Langmuir, in his desire to maintain the octet rule, was willing, in spite of the physico-chemical evidence, to postulate ionic structures for many of these species, whereas Lewis, in his desire to maintain the supremacy of the 2c-2e covalent bond, was willing to propose both hexet and expanded octet structures. The current consensus maintains both the octet rule, on the one hand, and the presence of covalent bonding, on the other, by abandoning the explicit assumption of both Langmuir and Lewis that the covalent bond must at all times be a localized two-centered bond. Thus electron deficient species have been accommodated via the closed 3c-2e bond popularized by Lipscomb for the boron hydrides (41) or via the use of delocalized π -electron systems (e.g., BF₃, CO₃²⁻, etc.), whereas electron-rich species (e.g., PCl₅, XeF₄) have been accommodated via the open 3c-4e bond popularized by Rundle (42), Pimentel (43), and others (44, 45).

The case of the *d*-block elements is more ambiguous. The generalized rare gas rule first hinted at by Langmuir was independently formulated by the English chemist N. V. Sidgwick in 1923 (46). Sidgwick, however, preferred to use total electron counts rather than valence electron counts and consequently called his technique the effective atomic number rule (EAN). Not until 1934 did Sidgwick explicitly connect his EAN rule with Langmuir's earlier 18-electron rule and derive an equation, related to Langmuir's (though based on total electron counts), for predicting the structures of polynuclear metal carbonyls and nitrosyls (47). Extensive applications of the EAN rule to the structures of metal carbonyls were made by Blanchard in the 1940's (48) and the importance of the rule exploded along with the field of organometallic chemistry in the late 1950's and 1960's, despite its many violations. More recent theoretical work has provided both quantitative (49) and qualitative (50) rationales for why the 18-electron rule is weaker than the octet rule.

Even Langmuir's arithmetical equation is still with us and

in modified form is used extensively in the field of solid-state chemistry. If one applies eqn. (3) to only the more electro-negative or anionic components of a solid-state compound, one obtains

$$\Sigma e_a = 8n_a - 2B_a \quad (5)$$

where Σe_a includes not only the valence electrons from the anionic components but any electrons transferred from the cationic components, n_a is the number of anionic components per unit formula, and $2B_a$ the number of anion-anion bonding electrons. In general the anionic components are restricted to elements lying to the right of the Zintl line in the periodic table (i.e., groups IV–VII, again shades of Mendeleev's higher types). Rearrangement of the equation and redefinition of $2B_a = b_a$, gives

$$8 = \frac{\Sigma e_a + b_a}{n_a} \quad (6)$$

This is called the generalized 8 – N rule and is the subject of an extensive literature (51–56). The equation can also be applied to the structures of pure elements lying to the right of the Zintl line. Some example applications are shown in Table 8.

Extended versions of Langmuir's original equation have also appeared in the literature which incorporate the possibility of the multicentered bonds required for the description of both electron-deficient (57) and electron-rich (58) species.

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Table 8. Examples of the Generalized 8 – N Rule

Species ^a	Σe_a	b_a/n_a	$(\Sigma e_a + b_a)/n_a$	Structural Array of the Zintl Atoms
C	4	4	8	$^3[C_{4/4}]$
P	5	3	8	$^2[P_{3/3}]$ or P ₄ tetrahedron
Se	6	2	8	$^1[Se_{2/2}]$
Cl	7	1	8	Cl ₂ discrete pairs
CsSn	5	3	8	(Sn ₄) ⁴⁻ tetrahedra
CaC ₂	10	3	8	(C≡C) ²⁻ triple bonded discrete pair
CaSi ₂	10	3	8	$^2[Si_{3/3}]^{1-}$
LaP	6	2	8	$^1[P_{2/2}]^{1-}$
CaP	7	1	8	P ₂ ²⁻ discrete pairs
NaI	8	0	8	isolated I ⁻

^a Anionic Zintl atoms in italics.

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