Two Conceptions of the Chemical Bond

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In this article I sketch G. N. Lewis's views on chemical bonding and Linus Pauling's attempt to preserve Lewis's insights within a quantum-mechanical theory of the bond. I then set out two broad conceptions of the chemical bond, the structural and the energetic views, which differ on the extent in which they preserve anything like the classical chemical bond in the modern quantum-mechanical understanding of molecular structure.

1. Introduction. Chemical bonds are central to understanding how matter behaves. The starting point of chemical explanation is a molecular structure for the relevant substance, a molecular structure just being atoms linked together in a certain way, by bonds. Chemical reactions are understood in terms of the breaking and making of bonds. Molecular spectra are explained as arising from the vibrations and rotations of bonds. From the middle of the nineteenth century, chemists began to explain the chemical behavior of substances in terms of structural formulas. They were not, at first, understood as embodying hypotheses about how the atoms are arranged in space, but instead as encoding a substance's patterns of chemical reaction or, perhaps less cautiously, the topological connections between its atoms. With the rise of stereochemistry in the last quarter of the nineteenth century came a fleshing out of structural formulas and a change in their status. Yet the physical nature of the bonds that held these structures together was a mystery.

It might be expected that the detailed application of quantum mechanics to chemical bonding, beginning in the second quarter of the twentieth century, would have resolved that mystery, but the need for approximate methods greatly complicates the explanatory relationship (see Hendry 2004). Quantum mechanics treats molecules as systems of electrons and

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nuclei interacting electrostatically. This determines a Schrödinger equation for the molecule whose solutions (molecular wave functions) correspond to the quantum states available to the system. But Schrödinger equations for chemically interesting molecules are mathematically intractable, and from the 1930s onward chemists developed approximate and semiempirical methods for solving them. Although the approximate solutions could be interpreted in terms of classical bonds, there was some question whether the bonds were projected into a quantum-mechanical reality that is devoid of them. The recovery of bonds, it was suggested, was an artifact of the approximate methods. To this situation there are two possible responses. One might regard the great unifying explanatory power of classical chemical structure, to which bonds are central, as an argument for the reality of bonds. If the interpretation of semiempirical wave functions in terms of bonds amounts to projection, this shows only that exact quantum mechanics alone is insufficient to capture the explanatory power of classical molecular structure. This is roughly the position adopted by Linus Pauling. A more revisionist alternative either eliminates bond talk altogether or, less radically, identifies bonds with something less problematic in physical terms. Since the role of bonds is to explain stability, the obvious choice is energy. This identification seems, however, to be highly revisionary of this central chemical concept.

In what follows I will first examine G. N. Lewis's views on bonding, as the fully developed conception of the bond prior to quantum mechanics. Then, in Section 3, I will briefly explain Linus Pauling's development of the semiempirical valence-bond method for calculating molecular wave functions, the skepticism about his methods, and his response to them. Finally, in Section 4, I set out two conceptions of the bond that (respectively) reject and embrace major revision of the classical bond: the structural and energetic views.

2. Lewis on Valence Formulas and Their Significance. In his influential book *Valence* ([1966] 1923), G. N. Lewis discussed the opposition between two great theories of chemical affinity of the nineteenth century (Lewis [1923] 1966, 20). In the electrochemical theory, affinity arises from the transfer of electricity between atoms: attraction between the resulting opposite charges explains the stability of the compound. As Lewis pointed out ([1923] 1966, 20), this account found support in the link that electrolysis established between electricity and chemical combination, but foundered on the existence of homonuclear species like H_2 and N_2. Also there were pairs of analogous compounds like acetic acid (CH_3-COOH) and trichloroacetic acid (CCl_3-COOH) in which positive hydrogen in one compound is substituted by negative chlorine in the other, without any great difference in chemical and physical properties, at least in ways that

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were detectable in the first half of the nineteenth century (for fuller details, see Brock 1992, Chapter 4).

The structural tradition, which had its roots in organic chemistry, was more diffuse. Rather than offering a precise physical realizer for chemical combination, it developed an important theoretical role for the bond as a structural feature of molecules. From the 1850s onward, chemists began to represent the chemical and physical behavior of organic substances using structural formulas in which the numbers of lines connecting the different atoms stood for their valences: their capacities to combine with fixed numbers of atoms of other elements. Such valence formulas were not at first taken to represent the real spatial positions of atoms within molecules, but their interpretation was enriched by the development of stereochemistry, a field in which there were intrinsically spatial theories of molecular structure whose explanatory power depended on their describing the arrangement of atoms in space. In the 1870s, Jacobus van 't Hoff explained why there are two isomers of compounds in which four different groups are attached to a single carbon atom by supposing that the valences are arranged tetrahedrally (the two isomers are conceived of as the mirror images of each other). Adolf von Baeyer explained the instability and reactivity of some organic compounds by reference to strain in their molecules (see Ramberg 2003, Chapters 3 and 4). Stereochemical explanation in organic chemistry was so successful that by 1923 Lewis was able to claim that "the valence of an atom in an organic molecule represents the fixed number of *bonds* which tie this atom to other atoms. Moreover in the mind of the organic chemist the chemical bond is no mere abstraction; it is a definite physical reality, a something which binds atom to atom. Although the nature of the tie remained mysterious, yet the hypothesis of the bond was amply justified by the signal adequacy of the simple theory of molecular structure to which it gave rise" ([1923] 1966, 67). Lewis was fulsome in his description of the unifying explanatory power of structural formulas: "No generalization of science, even if we include those capable of exact mathematical statement, has ever achieved a greater success in assembling in simple form a multitude of heterogeneous observations than this group of ideas which we call structural theory" (Lewis [1923] 1966, 20–21). But in this very versatility and explanatory power there was the danger of overextending the theory:

> The great success of structural organic chemistry led to attempts to treat inorganic compounds in a similar manner, not always happily. I still have poignant remembrance of the distress which I and many others suffered some thirty years ago in a class in elementary chemistry, where we were obliged to memorize structural formulae of a great number of inorganic compounds. Even such substances as the

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> ferricyanides and ferrocyanides were forced into the system, and bonds were drawn between the several atoms to comply with certain artificial rules, regardless of all chemical evidence. Such formulae are now believed to be almost, if not entirely, devoid of scientific significance. ([1923] 1966, 67)

Referring to Alfred Werner's critique of valence formulas, Lewis points out that "Werner does not distinguish between polar and non-polar compounds and therefore argues against the valence formula in general, but it will be noted that his arguments are based upon examples chosen from among polar compounds only" (1913, 1453). This distinction was central to Lewis's own view of the chemical bond. Taking potassium chloride and methane as examples of polar and nonpolar substances, respectively, in 1913 he argued that "we must recognize the existence of two types of chemical combination which differ, not merely in degree, but in kind" (1913, 1448). Nonpolar compounds are "immobil," that is, "unreactive, inert and slow to change into more stable forms, as evidenced by the large number of separable isomers. Inorganic compounds, on the other hand, approach more frequently the ideal polar or mobil type, characterized by extreme reactivity" (1913, 1448–1449). This distinction is "roughly" but not exactly coextensive with that between inorganic and organic chemistry (1916, 764). It is also relational, since it depends on the environment: a nonpolar substance may be polarized by a polar solvent (1916, 765). The differences are accounted for by different bonding in the two kinds of cases.

> To both types of compounds we should ascribe a sort of molecular structure, but this term doubtless has a very different significance in the two cases. To the immobil compounds we may ascribe a sort of *frame* structure, a fixed arrangement of the atoms within the molecule, which permits us to describe accurately the physical and chemical properties of a substance by a single structural formula. The change from the non-polar to the polar type may be regarded, in a sense, as the collapse of this framework. The non-polar molecule, subjected to changing conditions, maintains essentially a constant arrangement of the atoms; but in the polar molecule the atoms must be regarded as moving freely from one position to another. (Lewis 1913, 1449)

Although Lewis somewhat overemphasizes the contrast, and its alignment with the distinction between polar and nonpolar compounds, one consequence of the mobility of the polar compounds is that they exhibit tautomerism, where different molecular structures exist in "mobil equilibrium" and so "the compound behaves as if it were a mixture of two

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different substances" (1913, 1449). More than one tautomeric structure is required to account for the chemical and physical behavior of the whole population, but as Lewis points out in a later paper, "we must not assume that all of the molecules of the [tautomeric] substance possess either one structure or the other, but rather that these forms represent the two limiting types, and that the individual molecules range all the way from one limit to the other" (1916, 776). By noting that tautomerism is characteristic of polar compounds "we may account for the signal failure of structural formulae in inorganic chemistry" (1913, 1449). Organic substances are sometimes polar too, so tautomerism arises also in organic chemistry (in fact, modern chemistry associates the term primarily with that field), but it is more characteristic of 'immobil' nonpolar compounds to exhibit isomerism, where transition between the two forms is restricted, and they can be separated as distinct substances.

This is one respect in which Lewis considers valence formulas to be representative of real structure only for nonpolar compounds. Another important respect concerns the directionality of bonding. Lewis (1913, 1452) considers a proposal to represent the polar bond in potassium chloride with an arrow, as $K \rightarrow Cl$, to signify that "one electron has passed from K to Cl," but considers it misleading, because even if one could track the electron as it passed from K to Cl, the bond does not arise from this passage, for "a positive charge does not attract one negative charge only, but all the negative charges in its neighbourhood" (1913, 1452). In a nonmolecular polar substance like potassium chloride, the bonding is electrostatic and therefore radially symmetrical. Hence an individual ion bears no *special* relationship to any one of its neighbors. In short, the polar bond is nondirectional.

Lewis offered the pairing of electrons as a unifying explanation of bonding in both polar and nonpolar compounds. Bonds arise from atoms with incomplete electron shells filling them either by sharing electrons (in covalent, or shared-electron bonds) or transferring them (the ions and electrostatic bonding of polar compounds). Because shared electrons may not be shared equally (so that there are partial charges on the bonded atoms), Lewis saw pure covalent and ionic bonds as two ends of a continuum, thus presenting his theory as a synthesis of the dualistic and structural theories. Some features of his account were short-lived: he represented the fact that filled outer shells of atoms contained eight electrons with static arrangements of electrons at the corners of cubes. But his representation of electrons as paired dots (or colons) was applied immediately by organic chemists seeking to understand the mechanisms of organic reactions (see Brock 1992, Chapter 13). What is most important for this article, however, is Lewis's subtle understanding of the limited scope of the structural interpretation of the chemical bond. As relatively

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discrete structural features of molecules, bonds are to be found only where valence formulas are structurally significant.

3. Enter Quantum Theory. Linus Pauling's rise to prominence was closely associated with the application of quantum mechanics to explain chemical bonding. In 1928 he published a presentation for a chemical audience of the methods used by Walter Heitler and Fritz London on the hydrogen molecule (Pauling 1928b) and a brief note relating them to G. N. Lewis's theory of bonding, sketching how they might be extended to account for the tetrahedral structure of methane (Pauling 1928a). In a series of papers starting in 1931 he developed a quantum-mechanical theory of bonding that was only partially grounded in quantum mechanics, because it used approximations that were justified by detailed calculations only for diatomic molecules but were applied more generally to polyatomic molecules (Park 2000, Section 4). The origins of this work lay partly in physics. In a pioneering paper on many-body quantum mechanics, Werner Heisenberg had represented the two-electron states of the helium atom as superpositions of degenerate states, each corresponding to products of one-electron states. The term 'resonance' arose from the formal analogy between quantum-mechanical degeneracy and the interactions of coupled oscillators. Heitler and London had treated the hydrogen molecule in the same way, with a superposition of two equivalent states in which the electrons are assigned to the atomic orbital of first one hydrogen atom, then the other. Pauling's contribution was to tie these methods to the static electronic structures of G. N. Lewis (see Pauling 1928a, 359–360; Park 1999, 24–28), interpreting a molecule's quantum-mechanical states as superpositions of classically bonded structures. Pauling emphasized the autonomy of the chemical applications of resonance:

> The theory of resonance should not be identified with the valence-bond method of making approximate quantum-mechanical calculations of molecular wave functions and properties. The theory of resonance is essentially a classical chemical theory (an empirical theory, obtained by induction from the results of chemical experiments). Classical structure theory was developed purely from chemical facts, without any help from physics. The theory of resonance was also well on its way toward formulation before quantum mechanics was discovered. . . . The suggestion made in 1926 by Ingold and Ingold that molecules have structures that differ from those corresponding to a single valence-bond structure was made on the basis of chemical considerations, without essential assistance from quantum mechanics. It is true that the idea of resonance energy was then provided by quantum mechanics . . . but the theory of resonance has gone far

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> beyond the region of application in which any precise quantum mechanical calculations have been made, and its great extension has been almost entirely empirical with only the valuable and effective guidance of fundamental quantum principles. (Pauling 1956, 6–7)

The link between resonance and classical structure theory meant that the inscrutable notion of quantum-mechanical resonance was embedded in a theory of bonding that was understandable to chemists unfamiliar with the mathematical details of quantum mechanics.

Pauling's efforts to accommodate chemical intuition were not without their critics. For Robert Mulliken, Pauling was a mere "showman" whose deceptively simple explanations were "crude" though "popular" (Mulliken, quoted in Park 1999, 28). Mulliken worried that Pauling's valence-bond explanations were too quick and that, by appealing to arbitrary features of an approximate mathematical scheme, they threatened to set back the new discipline of quantum chemistry. The explanatory status of resonance was questioned even by proponents of the valence-bond approach. G. W. Wheland argued that resonance was a "man-made" artifact:

> Resonance is not an intrinsic property of a molecule that is described as a resonance hybrid, but is instead something deliberately added by the chemist or physicist who is talking about the molecule. In anthropomorphic terms, I might say that the molecule does not know about the resonance in the same sense in which it knows about its weight, energy, size, shape, and other properties that have what I might call real physical significance. Similarly . . . a hybrid molecule does not know its total energy is divided between bond energy and resonance energy. Even the double bond in ethylene seems to me less "man-made" than the resonance in benzene. The statement that the ethylene contains a double bond can be regarded as an indirect and approximate description of such real properties as interatomic distance, force constant, charge distribution, chemical reactivity and the like; on the other hand, the statement that benzene is a hybrid of the two Kekulé structures does not describe the properties of the molecule so much as the mental processes of the person who makes the statement. (Wheland, letter to Pauling, quoted in Simoes and Gavroglu 2001, 65)

But Pauling did not agree that bonds and resonance could be separated in this way: "You mention that if the quantum mechanical problem could be solved rigorously the idea of resonance would not arise. I think that we might also say that if the quantum mechanical problem could be solved rigorously the idea of the double bond would not arise" (Pauling, letter to Wheland, quoted in Park 1999, 43).

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For Pauling, the double bond and benzene's resonance structures were intimately connected: if one was "man-made" then both were. Pauling was awarded the Nobel Prize in Chemistry in November 1954, and in his Nobel lecture (Pauling 1964) he defended resonance in two ways. The theory had been the victim of an "unnecessarily strong emphasis on its arbitrary character" (1964, 433), and his first defense was consequentialist: the "convenience and usefulness are so great as to make the disadvantage of the element of arbitrariness of little significance" (1964, 433). The second defense underlines his response to Wheland: idealization is present also in classical structure theory, central to which is the idea that different molecules are assembled from the 'same' stock of bonds and functional groups. Each theory appeals to "idealized, hypothetical structural elements" (1964, 433). Although the real structure of benzene corresponds to none of the idealized Kekulé structures that the theory of resonance employs to describe it, the propane molecule likewise "has its own structure, which cannot be exactly composed of structural elements from other molecules" (1964, 433–434). Propane cannot contain any part that is "identical with a portion of the ethane molecule" (1964, 434), and so the similarities between organic molecules on which structure theory depends for both classification and explanation are idealized.

Pauling appealed to the linkage between resonance and classical structure in later defenses of the concept of resonance. In the third (1960) edition of *The Nature of the Chemical Bond* he incorporated a chapter titled "Resonance and its Significance for Chemistry," in which he emphasized its continuity with the classical notion of tautomerism. There was "no sharp distinction" between the two: the difference was a matter of the degree of the stability of the individual valence-bond structures (Pauling 1960, 564). In a retrospective article published in 1970, Pauling once again defended resonance by its connection to classical structural theory: "Bonds are theoretical constructs, idealizations, which have aided chemists during the past one hundred years in developing the convenient and extremely valuable classical structure theory of organic chemistry. The theory of resonance constitutes an extension of this theory. It is based upon the use of the same idealizations—the bonds between atoms—as used in classical structure theory, with the important extension that in describing the benzene molecule two arrangements of these bonds are used, rather than only one" (Pauling 1970, 999).

Classical structures and bonds are well supported by a wide variety of chemical and physical evidence. Resonance theory is independently supported by its explanatory successes as a "new semi-empirical structural principle" that is "compatible with quantum mechanics and supported by agreement with the facts obtained by experiment" (Pauling 1970, 1001).

But the linkage between resonance and bonds could cut both ways. If

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bonds are mere idealizations, and cannot be grounded in exact quantum mechanics, then perhaps they ought to lose their role in chemical explanation. Charles Coulson expressed this skeptical attitude as follows: "From its very nature a bond is a statement about two electrons, so that if the behaviour of these two electrons is significantly dependent upon, or correlated with, other electrons, our idea of a bond separate from, and independent of, other bonds must be modified. In the beautiful density diagrams of today the simple bond has got lost" (Coulson, quoted in Simoes and Gavroglu 2001, 69).

4. Two Conceptions of the Chemical Bond. In this section I articulate two conceptions of chemical bonding that express the respective intuitions of each side of the debate outlined in Section 3: Pauling's defense of his approximate methods, and the skepticism of Wheland and Coulson.

4.1. The Structural Conception. The core intention of the structural view of the bond is to retain the explanatory insights afforded by classical structural formulas. Lewis and Pauling work within this view, but the theoretical role of bonds within structural formulas predated both the discovery of the electron and the advent of quantum mechanics. Also, the view ought to be compatible with discoveries that have come after Lewis and Pauling. Hence it ought not to incorporate too closely the particular conceptions of the material basis of the bond offered either by Lewis (discrete shared electron pairs) or Pauling (hybridization and resonance). One way to meet these constraints is for the structural view to identify a theoretical function for bonds—continuous with that in the classical formulas—but leave it to empirical and theoretical investigation to identify their physical realizers within the quantum-mechanical states of molecules. Hence, on the structural view, chemical bonds are, at least for molecular substances, material parts of the molecule that are responsible for spatially localized submolecular relationships between individual atomic centers.

There are two sorts of challenge to the structural view: quantum-mechanical holism and electron delocalization, and the substances whose structures cannot informatively be represented by classical valence formulas. The holism objection is that quantum mechanics requires molecular wave functions to be symmetrical with respect to electron permutation. If so, features of molecular wave functions cannot depend on the identities of particular electrons. Understood as pairs of electrons with fixed identities, bonds simply cannot be features of molecular wave functions. A related problem concerns electron delocalization: electron density is 'smeared out' over the whole molecule, and so Lewis-style bonds consisting of pairs of electrons held fixed between pairs of atoms are not to

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be found. These two problems rule out the identification of bonds with Lewis-style static electron pairs, but they can be accommodated within the structural view. The first problem can be addressed by recognizing that a bond must be individuated by the atomic centers it links. Insofar as electrons participate physically in the bond (as they must), they do so not as individuals but as the occupancies of nonarbitrary partitions of the full electronic wave function that can be associated with the bond. The second problem can be addressed in similar fashion: some part of the total electron density is responsible for the features associated with the bond, and it will be a matter of empirical and theoretical investigation to identify which part. The structural conception is the heir to the conception of bonds that Lewis inherited from the structure theory of the nineteenth century, but generalized to take account of quantum-mechanical insights.

Turning now to the second objection, many substances seem to defy informative representation by valence formulas. Some, like BF_3, PF_5, and SF_6, violate the octet rule, according to which the outermost shell of a bonded atom has eight electrons. Such cases were known to Lewis and constitute objections to the octet rule rather than to the electron-pair bond. For other substances like the boranes (boron hydrides), although Lewis-style valence structures can be drawn (albeit violating the octet rule), these do not reflect known features of their structures like bond lengths and bond angles, which seem to demand distributed multicenter bonds (see Gillespie and Popellier 2001, Chapter 8). The response to these difficulties is to take a lead from Lewis, who, as we saw in Section 2, recognized that even where they can be written, classical structural formulas are not always structurally representative. If we regard the concept of a covalent bond as a theoretical notion associated with structural formulas, the applicability of this notion must be delimited by the applicability of structural formulas themselves. Of course there is *bonding* in paradigm ionic substances like potassium chloride, but their structure and stability must be explained without recourse to spatially localized submolecular relationships between individual atomic centers. The structural view maintains, however, that a proper understanding of the structure and stability of paradigm nonpolar substances like methane, to which structurally informative valence structures can be given, must involve bonds, understood as localized and directional submolecular relationships between individual atomic centers.

4.2. The Energetic Conception. The energetic conception of the bond is the logical outcome of Coulson's skeptical thoughts about the bond and the breakdown of valence formulas in many compounds. Rather than seeking a material element that realizes the theoretical role of keeping a

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molecule together, the energetic conception fixes on what is common to all cases of chemical bonding: changes in energy. On the energetic view, facts about chemical bonding are just facts about energy changes between molecular or supermolecular states. There is no requirement, or motivation, for bonds to be localized or localizable within the molecule, or directional. Hence the energetic view is more general and agnostic than the structural view and is more a theory of chemical bonding than a theory of bonds.

The energetic view finds support outside quantum mechanics. In thermodynamics, the strength of bonds can be estimated using Hess's law, according to which the change in enthalpy between two states is independent of the specific path taken between them. A measure of the strength of the carbon-hydrogen bond in methane, for instance, can be estimated by breaking the process of the formation of methane down into formal steps: the atomization of graphite and molecular hydrogen followed by the formation of methane. The heat of formation of methane from graphite and molecular hydrogen, and also the heats of atomization of graphite and molecular hydrogen, are empirically measurable, and the energy change in the formation of the C-H bonds is just the difference between them. In similar fashion, the lattice energy of common salt is the change in enthalpy when the salt lattice is formed from the gaseous ions Na^+ and Cl^-.

Within quantum mechanics, the molecular orbital approach represents the formation of a bond by correlating the electronic configuration of the molecule's bonded state with that of the separated atoms. A bond is formed just in case the bonded state is of lower energy than the separated atoms. The electronic configuration is described in terms of delocalized molecular orbitals, spread over the entire molecule, and the molecular geometry is explained as a local minimum on a potential energy surface, determined by the dependence on geometry of the occupied orbital energies.

While it has the advantage that it applies straightforwardly to all types of bonding, and is clearly consistent with quantum mechanics, the cost of this view is the loss of what is plausible and intuitive in the structural bond in the cases where valence formulas are informative. Radical revision of the classical conception of molecular structure implies explanatory loss.

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