Some comments on Linnett Double Quartet theory

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The recent critical evaluation of Linnett Double Quartet theory by Langler, Trenholm, and Wasson is critically examined and found to be based on misrepresentations of the theory's basic postulates and its use in predicting chemical reactivity. Various ways in which Linnett's original work can be profitably extended are suggested and briefly discussed.

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Un examen approfondi révèle que la critique récente de la théorie du double quartet de Linnet faite par Langler, Trenholm et Wasson est le reflet d'une mauvaise interprétation des postulats de base de la théorie et de leur utilisation dans la prévision de la réactivité chimique. On suggère différents moyens d'utiliser avec profit le travail original de Linnet et on en discute brièvement.

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The April 15, 1980, issue of this journal carried an article by Langler, Trenholm, and Wasson (1) which purports to be a critical reexamination and evaluation of the application of Linnett Double Quartet (LDQ) theory to problems of structure, reactivity, and resonance in organic chemistry. This article presents an interpretation of LDQ theory which is, in the present writer's opinion, incorrect and potentially misleading for those readers who are only superficially acquainted with Linnett's work. Consequently, it is felt that some comments on this interpretation are in order. On the other hand, the article does call attention to a number of situations which, though far from invalidating LDQ theory as the authors contend, do point to a need for a more explicit consideration of nuclear–nuclear repulsions in certain qualitative applications of the theory. Such a treatment is made in the present paper and is shown both to resolve the problems raised by Langler *et al.* and to point to a new area of application for Firestone's theory of Linnett or L strain (2, 3).

The Role of the Nuclei in LDQ Theory

The major source of difficulty with the authors' interpretation of LDQ theory stems from their misconceptions about its primary postulates. After first describing the concept of two tetrahedral spin sets, the authors summarize the theory as follows: "Molecular structures are elaborated by maximizing electronic separations and minimizing electronic repulsions. Priority, it is assumed, must be given to maximizing the separation of electrons having the same spin." This is correct as far as it goes, but appended to this statement is a footnote which reads, "The role of the nuclei is ignored." In other words, the authors claim that LDQ theory is based on the premise that electron correlation is the *sole* determinant of both molecular structure and

reactivity. This is simply not true. Thus, for example, Dale (4) in his summary of LDQ theory lists, in addition to the postulates dealing with electronic charge and spin correlation, the following two additional postulates. (*1*) "The attraction between electrons and atomic nuclei tends to draw a maximum number of electrons to regions between atoms." (*2*) "Two electrons of opposite spin may occur together as a pair (coincidence) when more electrons can thereby be concentrated between two atoms to form a bond." Likewise, Lawless and Smith (5) state the underlying principles of LDQ theory to be that "electrons are electrostatically attracted by the nuclei, are mutually repelled by other electrons, and have a tendency to achieve a fairly uniform charge distribution in a given chemical species, as limited by the effect of the spin wave function and relative electronegativities of the atoms involved."

In both statements the nuclei are assigned a key role. Structures are a result of the dynamic interplay between the desire of the electrons, on the one hand, to reduce their mutual repulsions by maintaining their spin sets and as much anticoincidence between the sets as possible, and the desire of the nuclei, on the other hand, to increase their electron attractions and reduce nuclear–nuclear repulsions by concentrating as many electrons as possible along the internuclear axes. Indeed, the role of the nuclei is assumed to be so apparent from traditional bonding concepts that the two standard books on LDQ theory by Luder (6) and Linnett (7) do not even explicitly state postulates dealing with these effects, the authors concentrating instead on the novelty which the postulate of spin correlation introduces into the treatment of the electron repulsions. Nevertheless, a reading of these books quickly shows that the nuclei play the roles outlined by Dale, and by Lawless and Smith.

Linnett himself originally presented LDQ theory as a simple modification of Lewis' original bonding concepts (8). Like both the two-dimensional topological Lewis structures themselves, and their three-dimensional extensions in terms of the tangent sphere model (9–14) and the VSEPR rules (15), LDQ theory makes use of such concepts as the octet rule (for period 2 elements), the electroneutrality principle (in terms of favorable formal charge distributions), and the isoelectronic principle, as well as assorted electrostatic arguments common to all such localized bonding models. Its sole innovation is its use of the postulate of the two tetrahedral spin sets to account for the effects of the Pauli principle and its attempt to maximize their degree of correlation within the bounds set by the other electrostatic energy components. The practical consequences of this are the appearance of valence electrons which are not close-paired and the possibility of one, three, and five electron bonds in addition to the close-paired two, four, and six electron bonds of the conventional Lewis – tangent sphere – VSEPR model. As Serratosa has succinctly put it, "All fundamental principles of chemical bonding are preserved in Linnett's theory, except the 'idolatry' for the Lewis *doublet*" (16).

It is true, however, that writers on LDQ theory, while always emphasizing the role of the nuclei in forcing the spin sets into coincidence, have often had little or nothing to say about the relative efficiency of various electron configurations in reducing nuclear–nuclear repulsions or about the relative efficiency of various kernels, as expressed by their charges and sizes (i.e., electronegativities), in forcing the electrons into coincidence and in distorting the spin sets due to inequalities in their relative attractions for the electrons. This emphasis (or lack thereof) is in fact consistent with the origins of LDQ theory as, like the Lewis – tangent sphere – VSEPR model, it assumes that electronic repulsions and electron–nuclear attractions are the two major determinants of molecular structure in the case of primarily covalent species. However, in so doing, LDQ theory does not deny the existence of nuclear–nuclear repulsions nor that they play a role in determining the finer details of structure and reactivity (or, indeed, a major role in the case of ionic species). Bent, for example, has made a number of specific applications of nuclear–nuclear repulsions within the context of LDQ theory in order to rationalize certain trends in bond angles and bond lengths (9–14), and Firestone has called attention to a number of situations in which it is not possible to arrive at a satisfactory disposition of the tetrahedral spin sets without simultaneously moving bonding electrons away from the regions of the nuclear–nuclear axes (2, 3). This frequently occurs in the case of transition state species and the resulting destabilization, due to the increase in nuclear–nuclear repulsions and the decrease in electron–nuclear attractions, has been termed Linnett or L strain by Firestone and has been used by him to analyze the origin of activation energies in the case of E2, S_H2, and S_N2 reactions (17). A similar explicit treatment of nuclear–nuclear repulsions quickly resolves a number of the problems discussed by Langler *et al*.

A case in point is the authors' treatment of the excited states of oxygen. The Linnett structures of the ground state ($^3\Sigma_g^-$) and first two excited states ($^1\Delta_g$) and ($^1\Sigma_g^+$) are given by structures 1–3. The authors point out that the ground state 1 and the first excited state 2 differ very little in the degree of electron close-pairing and yet have an energy separation of 22.6 kcal/mol, whereas the first excited state 2 and second excited state 3 differ a great deal in the amount of electron repulsion, all six electron pairs having become close-paired in the $^1\Sigma_g^+$ state, and yet differ in energy by only 15.1 kcal/mol. They conclude that while an "analysis of apparent electronic separations is qualitatively helpful ... (it) ... can be somewhat misleading quantitatively." However, an examination of structures 1 and 2 shows that while both exhibit similar degrees of electron correlation and both place four electrons in the bond region, structure 1 concentrates at least one of these electrons directly on the nuclear–nuclear axis, whereas structure 2 does not. The resulting difference in the efficiency of screening the nuclear–nuclear repulsions, as well as the greater electron–nuclear attraction for the electron so placed, should account for most of the 22.6 kcal/mol separation. Structures 2 and 3, on the other hand, do not differ significantly in the degree to which they concentrate electrons on the nuclear–nuclear axis and their somewhat lower energy separation of 15.1 kcal/mol should be largely due to differences in electron repulsion energy resulting from the increased close-pairing in structure 3.

1   2

3
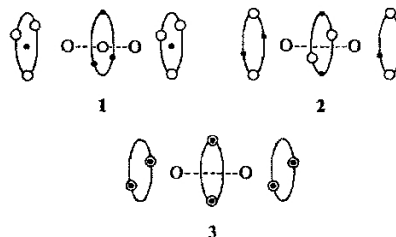
A similar analysis resolves the anomalies in the average bond energies per CC bonding pair discussed by the authors later in the paper (where we are again reminded in a footnote that the nuclei are neglected). Using Linnett structures 4–6 for

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5

6
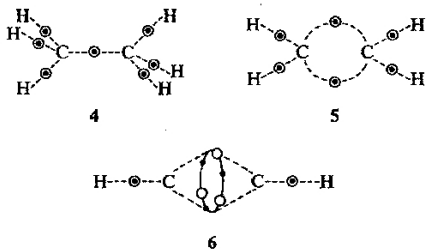
ethane, ethene, and ethyne, the authors argue that LDQ theory would predict a similar average energy per CC bond pair in ethane and ethene, as both contain close pairs, but a much higher energy per pair in ethyne where the pairs have achieved a degree of anticoincidence. They then point out that the observed energies per pair are exactly the opposite, being 83.1 kcal/mol for ethane, 73.5 kcal/mol for ethene, and 64.6 kcal/mol for ethyne. Again, as with the simplest possible electrostatic model, LDQ theory would not predict a double bond to be twice as strong as a single bond. The close pairs in ethene are not localized directly on the nuclear–nuclear axis like the pair in ethane and the resulting increase in nuclear–nuclear repulsion (and decrease in electron–nuclear attraction) should lead to a lower average energy per bond pair in ethene. In ethyne the nuclear–nuclear repulsions would be even greater as the spin set polyhedra now share common faces rather than common edges, drawing the nuclei even closer together without at the same time increasing the degree to which the electron density is directly concentrated on the nuclear–nuclear axis. However, the decrease in electron repulsion in ethyne should partly offset this increase and, if anything, the drop in energy per CC pair on going from ethene to ethyne should be somewhat less than the drop on going from ethane to ethene, as is in fact observed.

Molecular Structure

Given the authors' premise that LDQ theory denies the role of the nuclei in structure formation, it is difficult to imagine how they account for the electrons ever forming close pairs; indeed, in the case of simple hydrides they assume that the electrons don't. Any attempt to invoke close-pairing of the electrons due to electron–nuclear attractions is labeled as inconsistent. Instead the authors postulate that the central atoms in CH_4, NH_3, and H_2O maintain the completely correlated spin sets of Ne and they place the protons on lines connecting electrons of opposite spin with no electron density whatsoever directly between the proton and central kernel. The completely correlated electrons of Ne lie at the corners of a cube formed by the interpenetration of the two tetrahedral spin sets, electrons of like spin being separated by the face diagonals of the cube and electrons of opposite spin by the cube edges. Needless to say, given 12 edges and only four to two protons, several alternatives are equally likely, leading to the authors' claim that LDQ theory "may be ambiguous" in predicting molecular geometry. In the case of CH_4, for example, they conclude that, on the basis of electronic correlation alone, square planar and tetrahedral methane are equally likely, but by invoking proton–proton repulsions arrive at the tetrahedral structure. A similar procedure in the case of NH_3, however, leads to the incorrect conclusion that it is trigonal planar.

Surely, given the proton's size, no nucleus or kernel could be expected to be more effective in inducing close pair formation. Only such small highly-charged kernels as F^{7+} and O^{6+} would be capable of combating this tendency, as is postulated in the case of hydrogen-bonded species such as HF_2^-. These are the only cases in which Linnett appears to propose structures for stable long-lived species in which the electrons bonded to H are not close paired and even here at least one electron per bond is placed directly between the proton and the other kernel. Though the authors' treatment of hydride geometries may be viewed as an unsuccessful attempt on their part to try new variations of Linnett's postulates in which electron repulsions and nuclear–nuclear repulsions are considered as the major determinants of structure, rather than electron repulsions and electron–nuclear attractions, this treatment does not correspond to that of standard LDQ theory, which correctly predicts that NH_3 is pyramidal (6, 7).

However, the authors do call attention to a number of interesting problems relating to the structures of hydrocarbon anions. They point out, for example, that it is impossible to write a LDQ structure for the allyl anion in which one has both resonance stabilized CC bonds (i.e., two non-close-paired, three-electron CC bonds rather than one close-paired double and one close-paired single bond) and close-paired CH bonds at the same time. Using Linnett's more condensed symbolism, in which a thin line represents an anticoincident pair and a thick line a coincident or close pair, the struc-

ture for the allyl anion (which is isoelectronic with O_3) may be written as

7
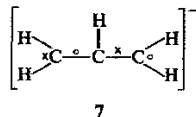
The authors also propose a LDQ structure for the ethene anion which suffers from the even graver problem of simultaneously forcing three electrons into spatial coincidence, two of which have the same spin

8
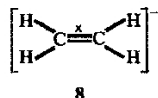
and they imply that this result casts severe doubts on the validity of standard LDQ theory. However, this proposed structure is incorrect on several counts. Not only does it violate the octet rule for carbon, it incorrectly predicts a bond order of $2\frac{1}{2}$ for the CC bond. Addition of an electron to the π^* LUMO of ethene should in fact reduce the CC bond order to $1\frac{1}{2}$. The proper Linnett structure for $C_2H_4^-$ (which is isoelectronic with O_2^-) should be:

9
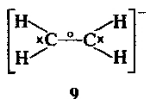
Unlike structure 8, this structure does not require the spatial coincidence of three electrons and gives the proper CC bond order. However, like the structure for the allyl anion, it leads to a lack of close-pairing in the CH bonds.[1]

What both the allyl and ethene anion structures appear to represent is, in fact, a previously unrecognized example of the application of Firestone's concept of Linnett or L strain (2, 3). Because it is not possible to reduce the electron repulsions in the CC bonds without simultaneously increasing the nuclear–nuclear repulsions in the CH bonds, these species suffer, like the transition states discussed

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[1]It should be noted that the authors themselves also suggest a resolution of the ethene and allyl anion structures which involves separation of the electrons in the σ bonds. However, they introduce a new symbolism into their formulas at this point and never explain what it means. That is, from their proposed formulas, one cannot tell whether they are postulating an angular splitting or a radial splitting of the σ bonds. If their proposed structures are identical with LDQ structures 7 and 9, then it is difficult to understand why they bring up structure 8 in the first place and why they fail to cite Firestone's work for a proper interpretation of the result within the context of standard LDQ theory.

by Firestone, from L strain. Consequently, LDQ theory would predict that they should be highly reactive, particularly with respect to oxidation, as is indeed the case.

Reactivity

The authors' treatment of the application of LDQ theory to reactivity is even more misleading. An analysis of this treatment is simplified by first defining some vocabulary. By *kind* of bond we will mean the kind of nuclei involved in the bond (e.g., H—H vs. O—H); by bond *multiplicity* we will mean the total number of valence electrons in the bond, and by bond *electron correlation*, the arrangement of these electrons. This latter factor is unique to LDQ theory and affects the bond energy both through changes in electron repulsion energies and changes in the relative screening of the nuclear–nuclear repulsions.
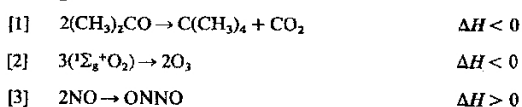
All of these factors affect the energy of a species and hence the energy of a reaction. Only in the case of a reaction in which the number, kind, and multiplicities of the bonds in the products and reactants are identical can one expect a correlation between ΔH of the reaction and the change in electron correlation. In cases in which one or more of the other factors also change, we can also expect a correlation if we compare two reactions such that the changes in all the factors, save the electron correlation, are the same for both reactions. In this case the correlation will involve the sign of the relative ΔH values (i.e., $\Delta\Delta H$) and the relative changes in the correlation energies for the two reactions; that is, it will tell us which of the two reactions is the more endothermic, but will tell us nothing about the absolute values or signs of the individual ΔH changes of each reaction.

A third situation in which LDQ theory may be applied is in predicting whether the experimental ΔH of a reaction will be more exothermic or more endothermic than the theoretical value calculated be means of simple Lewis structures and tabulated bond energies. This requires that LDQ theory predict the structure of either a single reactant or product to differ significantly from that given by its close-paired Lewis structure. This difference may involve both bond multiplicities and electron correlation. The situation is exactly similar to that used to evaluate resonance energies, the classic example being the difference between the experimental ΔH for the hydrogenation of benzene and that calculated for a single Kekulé structure. This analogy is more than formal as both resonance in VB theory and configuration interaction in MO theory are attempts, like the double-quartet hy-

pothesis, to account for electron correlation effects.

On the other hand, Langler *et al.*, consistent with their denial of the role of the nuclei, put forward the claim that LDQ theory pretends to be able to predict the sign of the enthalpy change for a reaction by simply evaluating the change in electron correlation, irrespective of the number, kind, and multiplicities of the bonds made or broken in the reaction. The procedure they adopt is to evaluate the change in the number of close pairs on going from reactants to products. If this change is positive (loss of correlation), ΔH is assumed to be positive; if it is negative (gain of correlation), ΔH is assumed to be negative. This procedure is then applied to ten reactions and shown to give an incorrect result for virtually all of them, allowing the authors to conclude that LDQ theory "does not provide a guide to chemical reactivity, in contrast to claims made for it." An examination of these reactions shows that eight of them involve the making and breaking of different kinds of bonds (mostly H—H bonds converted into O—H, N—H, and C—H bonds or vice versa) and that the remaining two involve changes in bond multiplicities. In short, none of them conform to the circumstances under which LDQ theory (in contrast to the authors) would postulate a correlation between ΔH and change in electron correlation.

It is instructive in this regard to examine the three reactions (not included in the table of ten just mentioned) which the authors quote[2] from Linnett's original paper (8) and book (7) in order to justify their procedure:

[1] $2(CH_3)_2CO \rightarrow C(CH_3)_4 + CO_2$ $\Delta H < 0$

[2] $3(^1\Sigma_g^+ O_2) \rightarrow 2O_3$ $\Delta H < 0$

[3] $2NO \rightarrow ONNO$ $\Delta H > 0$

Again making use of Linnett's condensed symbolism,[3] the first of these reactions may be written as


[4] 2
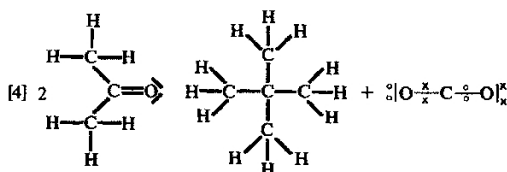
[2]See ref. 7, pp. 45–46 and ref. 8, p. 2647 and footnote 14 on the same page.

[3]Here we have positioned the lines for nonbonding pairs in a manner similar to conventional Lewis structures in order to avoid the appearance of dangling bonds conveyed by Linnett's original formulas.

As can be seen, the number, kind, and multiplicities of the bonds are the same on both sides. The correlation of the CC and CH bonds is also the same, only that of the CO bonds changing. In other words, unlike the ten reactions tested by the authors, this reaction conforms to the conditions under which there should be a correlation between ΔH and change in electron correlation. Interestingly, Linnett's purpose in quoting this reaction was not to explicitly predict the sign of its enthalpy change but to use its known ΔH value to estimate the energy involved per electron pair in close pairing.

The second reaction may be formulated as

[5]
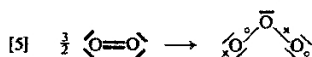
It fails to conform to the necessary conditions as there is a change in both bond multiplicity and electron correlation. However, Linnett does not pretend to predict the sign of ΔH for this reaction as the authors claim, but rather the sign of $\Delta \Delta H$ relative to the same reaction using the $^3\Sigma_g^-$ ground state of O_2

[6]
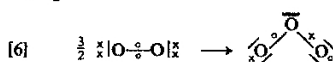
The change in bond multiplicity is the same for both reactions so $\Delta \Delta H$ must reflect the relative changes in the electron correlation energies for the two reactions. In keeping with the predictions of LDQ theory, reaction [5] is in fact found to be more exothermic than [6] by 57 kcal/mol. The fact that the ΔH of reaction [5] happens to be exothermic and that of [6] endothermic is an accident of the example used, and Linnett's correlation deals only with the sign of $\Delta \Delta H$ and not the signs of the individual ΔH values.

Finally, we have the third example, which involves changes in bond multiplicities, electron correlation, and kind of bond.

[7] $2 \times|N\overset{\circ}{=}O|\times \rightarrow$
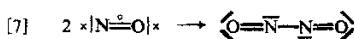
Here again Linnett is not predicting the sign of ΔH as the authors claim, but rather that it must be more endothermic than would have been predicted by the classically formulated version given by simple Lewis structures:

[8] 2 $\rightarrow$
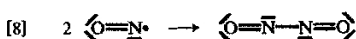
In other words, it is an example of the third situation discussed earlier.[4] Reaction [8] must be inherently exothermic as, like all simple radical dimeri-

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[4]A comparison of this type is also given for O_3 in ref. 7, pp. 61–62.

zations, it merely involves the formation of a new bond without any old bonds being broken in the process. In contrast, the Linnett structure in eq. [7] predicts that the N—N bond can only be formed at the expense of the multiplicity of the NO bonds and with loss of the electron correlation in the monomer. In other words, LDQ theory predicts that the experimental ΔH should be more endothermic than that predicted by eq. [8]. Linnett does not quantitatively test this prediction by comparing the experimental and calculated ΔH values, but instead remains content with the fact that NO doesn't dimerize to any great extent under ambient conditions, indicating that the endothermic shift ($\Delta\Delta H$) relative to the necessarily exothermic ΔH predicted by [8] is so large as to either make the real reaction inherently endothermic or the exothermic value so small that is is unable to offset the unfavorable entropy change expected for dimerization reactions in general.

Curiously, in their discussion of reactivity, the authors let pass without comment one of the few explicit attempts to apply LDQ theory to problems of reactivity, namely Firestone's (2, 3, 17) analysis of E2, S_H2, and S_N2 transition states in terms of Linnett or L-strain, even though they reference his papers on the subject. Finally, the authors' bibliography of the literature dealing with LDQ theory is far from complete, a number of major references being conspicuously absent (4, 5, 9–14, 18–39).

In conclusion, the paper by Langler *et al.* is anything but a critical evaluation of the application of LDQ theory to problems of structure and reactivity in organic chemistry. By denying the role of the nuclei and applying only the postulates dealing with electron repulsion, they produce a misleading picture of LDQ theory and then proceed to demonstrate that it leads to unsatisfactory results in predicting structure and reactivity. Indirectly, however, the Langler article points to the need for a greater development and refinement of Linnett's original work. One area of refinement, briefly touched on here, is a comparison of the relative efficiency of the electrons in bonds of the same kind and multiplicity, but differing correlation, in screening kernel–kernel repulsions.

A second area of refinement would be in assessing the relative efficiency of various nuclei or kernels in inducing close pairing. Other things being equal, one would expect this ability to increase with increasing kernel charge and decreasing kernel size. When kernels of identical charge but differing size act as central atoms in isoelectronic species, there will also be a tendency to resist ligand-induced close-pairing as the central kernel in-

creases in size. This is because the same number of valence electrons must screen a proportionately larger surface area in the case of a large kernel, or, conversely, close pairing for a large kernel will be more strongly resisted because it results in the descreening of a proportionately larger amount of surface area. Likewise, distortions of the shared spin sets due to differences in the electronegativities of the kernels, or even due to differences in the relative attraction of a kernel for the bonding versus the nonbonding electrons in a given spin set, can lead to a decrease in electron repulsion because of the resulting *radial* electron correlation, thereby supplementing the *angular* correlation which LDQ theory currently emphasizes.[5]

Considerations of this kind could easily provide a qualitative rationale for the discontinuity in structures and bonding properties shown on passing from period 2 species to their heavier group analogs (e.g., H_2O vs. H_2S, H_2Se, H_2Te; NH_3 vs. PH_3, AsH_3, SbH_3, etc.), accounting for the differences in bond angles, the inert pair effect, the reluctance of these elements to form multiple bonds among themselves, etc. Another area of application would be in assessing the relative amounts of L-strain to be expected in a series of S_N2 displacement reactions in which the donor species and acceptor atom are kept constant but the substituents on the acceptor atom are varied.

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