JOURNAL OF PHYSICAL ORGANIC CHEMISTRY
J. Phys. Org. Chem. 2005; **18:** 566–571
DOI: 10.1002/poc.909

Howard J. Lucas. A pioneer in the introduction of the teaching of physical organic chemistry at the college and university level of elementary organic chemistry

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Received 6 November 2004; revised 8 January 2005; accepted 8 January 2005

ABSTRACT: Virtually all recent textbooks for elementary organic chemistry courses now offer substantial components of physical organic chemistry, an emerging discipline first known by that name in 1940. It may come as a surprise to learn of an elementary textbook using many physical principles was published in 1935 by Howard J. Lucas of the California Institute of Technology. A comparison will be made of the subject matter of the Lucas book with two highly respected texts of about the same period, written by Paul Karrer and Louis and Mary Fieser.

KEYWORDS: Howard J. Lucas; teaching of physical organic chemistry; elementary organic texts; Paul Karrer; Louis Fieser; Mary Fieser

Before 1935, the textbooks of organic chemistry were generally descriptive of physical properties and reactions of carbon compounds, with some coverage of optical activity and geometric isomerism. The steps by which reactions occurred, in terms of the nature of possible intermediate substances, was a matter of speculation, but little if any of this involved physical chemistry principles, such as actual reaction kinetics, chemical thermodynamics and electronic theory. Indeed, organic and physical chemists usually regarded themselves as being in separate fields of chemistry, with walls fostered by ignorance of one another's fields and possibly with even suspicion of one or the other's activities.

Nonetheless, on each side of the divide, there was substantial experimentation and speculation about the properties of many organic compounds, especially where the properties did not fit easily into the conventional wisdom derived by the study of simple molecules. A prime example was benzene, discovered much earlier (1825) by Michael Faraday. Benzene, C_6H_6, is thermally stable and almost as unreactive as the so-called 'saturated' hydrocarbons of generic formula C_nH_{2n+2}. The Kekulé structure of benzene with its unsaturated double bonds linked by single bonds did not solve the problem of the low reactivity of benzene. It was recognized early on that the current conventional wisdom had to be updated and adjusted to the unusual lack of reactivity of benzene. This was initially achieved by evoking a new class of unreactive, unsaturated compounds called 'aromatic', of which there are now many examples, some of substantial complexity and many of commercial as well as biochemical importance. However, to evoke a new class of compound to account for a special stability is at best a palliative and an enormous effort was expended to rationalize this stability at a level suitable for use in an elementary organic course.

Explanation of the properties of aromatic compounds took an unusual confluence of individuals in different areas of chemistry, which helped to achieve a sea change in the relation between organic and physical chemists, and this catalyzed inclusion of physical organic principles in beginning organic chemistry. The California Institute of Technology played a very significant role in this process through the efforts and influence of a person barely known to currently active organic chemists, Howard J. Lucas[1] (1885–1963) (Fig. 1). Nonetheless, the 1935 Lucas textbook *Organic Chemistry*[2] was the first to incorporate physical organic chemistry as a major theme.

By training, Howard Lucas was not a person one would usually expect to have much influence in an area in which physical chemistry plays such an important role. A native of Marietta, Ohio, his early life was apparently that of a normal American boy, who attended Ohio State University, as both an undergraduate and graduate student and obtained a Master's Degree in 1908. His research was with William MacPherson, who was interested primarily in sugar chemistry and, for his thesis, Lucas worked on the reaction of 1-benzoyl-1-phenylhydrazine with *o*-benzoquinone. This work was published in the *Journal of the American Chemical Society* in 1909.[3]

It is a matter of record that Lucas started work with a fellowship towards his PhD at the University of Chicago,

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Figure 1. Howard J. Lucas, 1952. (Courtesy of John D. Roberts)

but after about 2 years, most likely facing financial responsibilities after the death of his father, he took a job with the US Department of Agriculture. He then assumed a position as chemist in the Puerto Rico Agricultural Experiment Station starting in 1910. During his 3 years stay in Puerto Rico, he published two papers, one on the analysis of the milk of Puerto Rican cows[4] and the other on a method of analysis for nitrobenzene in peanut oil.[5] The latter sounds like a no-brainer, but the fact is that nitrobenzene (a rather toxic substance) was cheaper than peanut oil and was commonly used in Puerto Rico as an adulterant of peanut oil.

With this background, there remains a substantial mystery as to how Lucas obtained a position at Throop College in 1913. Throop had been in existence for some years, but was not a school of special distinction. However, change was in the wind. George Ellerly Hale, the famous solar astronomer, builder of the Xerxes and Mount Wilson Observatories, along with Arthur Amos Noyes, a physical and inorganic chemist once President of MIT, and Robert Andrews Millikan, a renowned physicist, were planning to essentially take over Throop College and convert it into a world leader in scientific education and research as the California Institute of Technology. If so, one wonders why Noyes, if involved, would allow the organic chemistry staff to be a single person with such meager-sounding academic credentials. One possible hypothesis was that no one else wanted to take the job. Another seemingly implausible rumor was that Noyes, an inorganic chemist, wanted the future Caltech organic professor to be a person who would not burden the Division with a large research group, but would give solid organic chemistry lectures and a laboratory course for undergraduates.

If the latter were true, Lucas did much more than Noyes expected. The reason was partly that he had an inquiring mind and was also a gregarious person. But probably just as important, he was the only organic professor and, as time went on, he became surrounded by a coterie of world-class chemical luminaries, such as Linus Pauling, Arthur Amos Noyes, Richard Tolman, Richard Badger, Don M. Yost, Arnold Beckman and Roscoe Dickinson. If Lucas were to develop new insights and ways of teaching organic compounds and their reactions, he would best do it by drawing on the insight and expertise of his colleagues. This must have been the case, because in the preface of his organic text, he acknowledges the assistance of Linus Pauling, Don M. Yost and the later physical organic scholar and author George Willard Wheland (then a postdoctoral fellow at Caltech). Also included in the list is Charles D. Coryell, who was also a postdoctoral fellow, who worked with Pauling to demonstrate the paramagnetism of deoxyhemoglobin. Later Coryell, through his yeoman work in association with the Manhattan Project, where his group discovered the element promethium, became a professor of inorganic chemistry at MIT. Coryell's first academic position was as a laboratory instructor of physical chemistry at UCLA and inspired many students with his boundless interests and innovative ideas. Surely, he must have contributed much to the physical tone of the Lucas book.

So what was so unusual about the Lucas book? The truth is, a lot, and it should be remembered that Lucas published his book 5 years **before** Pauling's *Nature of the Chemical Bond*[6] and Hammett's immensely influential monograph *Physical Organic Chemistry*[7] appeared. What new ground did Lucas break? To answer that question, we need to compare the Lucas book with what was being covered by highly regarded elementary organic chemistry textbooks of the time. Here, we will look at two very widely used examples, one from Switzerland and one from the United States. *Organic Chemistry*[8] was written by Paul Karrer, an eminent Swiss chemist, who was awarded a Nobel Prize in 1937 for his studies of the naturally occurring carotenoids. The first edition of Karrer's book was published in German in 1928, went through many subsequent editions, and was essentially a bible of organic chemistry in Europe. The first English version came out in 1938. It was revised in 1942 and reprinted in 1946.

The second book, *Introduction to Organic Chemistry*,[9] was written by Louis and Mary Fieser, a well-known team, whose prose was so similar that it was virtually impossible to know which one wrote which sections or chapters. The first edition of the Fieser book came out in 1944. At that time, Louis Fieser was the Sheldon Emory Professor of Chemistry at Harvard and already the author of several books and very highly regarded for his research on polycyclic aromatic compounds. The justly famous Fieser and Fieser text was beautifully written and detailed in its descriptions of organic compounds and their reactions.

Initially, we will look at how each of these books outlines its goals and audience in its preface (throughout some abridgement, indicated by..., will be used to highlight the important points).

Karrer's preface in the 1946 English printing begins, 'The first German edition of *Lehrbuch der Organischen Chemie* appeared in 1928. The purpose and aim, which led me to write this textbook, have been kept in view throughout the later editions. This aim was to provide students with a textbook of organic chemistry of medium size, which would give them a survey of the ever-increasing body of facts. To make the problems of organic chemistry more easily understood, and to make the subject more real and live, special attention has been paid in all chapters to the description of methods of synthesis and of determining the constitution of organic compounds.... It is hoped that this textbook in its new form will prove a friend and a help to the student.'

An abridged preface of Fieser and Fieser, 1944 edition, follows: 'The main theme of this book is the development of the fundamental chemistry of the hydrocarbons, alcohols, acids, and other classes of organic compounds. Aromatic compounds are introduced at an early stage and illustrative examples from this field are used throughout, but the theory of the nature and substitution reactions of aromatic substances is reserved for specific discussion at a later point, after adequate groundwork has been laid. Thus, the empirical correlation of phenomena prevalent in the early chapters gradually gives place to interpretations in terms of modern theory.... The most novel feature of the book is the inclusion of a number of chapters for optional reading dealing with significant applications of organic chemistry to technology and to the biological and medical sciences.'

The Lucas preface of 1935 is excerpted here at much greater length, because he has so much to say that is directly relevant to the question of how physical organic chemistry first got into beginning organic textbooks. 'The subject of organic chemistry is so complicated and the reactions of organic compounds are so varied that oftentimes the student is submerged in a mass of detail and fails to grasp the underlying principles. As scientists learn more and more about the nature of matter and the manner in which atoms combine to form molecules, they are able to enunciate principles and laws. A satisfactory textbook of organic chemistry not only should present the subject matter in a systematic and logical manner, but also should correlate it with principles. In the attempt to do these things in this text, the author has adopted certain procedures, as follows: Emphasis has been placed upon class reactions, rather than upon the reactions of individual compounds; relationships with inorganic compounds are made use of, when possible; energy relationships are discussed and their significance pointed out; the underlying principles of molecular structure are early presented and applied throughout to individual cases; the electrochemical nature of radicals is indicated and their effect upon properties noted; and numerous applications of the electronic theory of valence are made.... In connection with energy relationships, the inclusion of a table of bond energies makes it possible to calculate approximate values for the heats of reaction of many organic reactions.... Important aspects of the electron theory are the types of valence bond, that is, whether it is an ionic, a covalent, a coordinate covalent, or a hydrogen bond; the possibility of resonance; and the influence of radicals upon electronic configurations. All of these factors are important, and their application to the problems of structure and properties will continue, as they have done in the past, to lead to a better understanding of organic chemistry.... It is felt that an understanding of the principles, which govern chemical and physical properties, is more important than an encyclopedic knowledge of chemical formulas and reactions. In keeping with this viewpoint, the problems have been devised to test the ability of students to think.'

Then, after suggesting how instructors might wish to organize a course based on the book, there is a paragraph which seems like an afterthought, but it is amazing to see it in a beginning organic textbook, even in 2004, 70 years later: 'The thermodynamic symbols used in heat content of the system, is expressed in kilocalories (i.e., kg. cal.), ΔF, the change in the free energy of the system, in calories, the three states of matter by (g), (l), and (s), and natural logarithms by ln. The heats of combustion given in the tables are kg. cal. per gram mole, and all temperatures without designation are degrees centigrade.' It is obvious from this quotation that introduction of physical principles was not going to be treated lightly in the subsequent presentation.

Next we will compare examples from the texts of Karrer, the Fiesers and Lucas with regard to the way they treat some simple organic fundamentals that are very basic to any plan to include physical organic concepts at the elementary level.

One set of concepts where any reasonably modern organic text must start was outlined many years earlier by G. N. Lewis with a very simple electronic theory of valence. The Lewis approach to chemical structures worked very well in both organic and inorganic chemistry. It was particularly useful in that, for normal compounds, it provided an understanding of simple chemical bonding by postulating that each neutral atom brought with it a specific number of valence electrons and formed single valence bonds by sharing one electron with an electron provided by another atom to form an electron-pair bond. Thus, a neutral nitrogen atom brings five electrons and is most stable with a filled shell of eight electrons. If it forms a shared bond with hydrogen, which when neutral has one electron, the hydrogen shell of electrons is full with just two electrons, to form N:H. However, when nitrogen is just sharing two electrons with hydrogen in N:H, it has only six electrons in it valence shell. In consequence, it can form two more shared electron bonds with two more hydrogens and thereby achieve a filled shell of eight electrons with the formation of NH_3, ammonia. Both nitrogen and each of the hydrogens in ammonia will be neutral and contributing five electrons and one electron, respectively, to the bonding. It will be seen that while nitrogen ends up with eight electrons in its valence shell, two electrons are left over in what is called an 'unshared electron pair'. This pair can be shared with a positive hydrogen (no electrons in its outer shell) to form ammonium ion, NH_4^+.

With this procedure and an electronically based periodic table, one can add up the number of electrons that each atom contributes in forming a molecule and then easily decide whether an overall chemical entity will be neutral or positively or negatively charged. Further, if the total number of valence electrons in the entity is an odd number, then the possibility of reactions that would cause the 'left-over' unpaired electron to become somehow lost, or else form an electron-pair bond, needs consideration. Compounds with an odd number of electrons are often more or less correctly known as 'free radicals'.

This type of aid to generalizing the conventional electron-pair bond structures was already beginning to penetrate into organic chemistry at the research level, but the use of Lewis structures in elementary organic texts was by no means extensive.

The Karrer text, despite its broad and deep coverage of organic compounds and reactions, barely mentions Lewis structures. To be sure, three pages were devoted to the electronic interpretation of the interaction of molecules having single and double bonds between their carbon atoms. Curiously, the exposition here uses x's to denote electrons, whereas later, in a half-page explanation of resonance in benzene, the conventional paired-dot symbols were employed.

Fieser and Fieser's *Introduction to Organic Chemistry*[9] was published in 1944, 9 years after Lucas, and has a rather light sprinkling of Lewis electron-pair structures, some in brief discussions of resonance theory. One can be surprised at Louis Fieser, because he took his PhD on the study of reduction potentials of quinones with James Bryant Conant, an early, very capable physical organic chemist. To be fair, the Fiesers, as we shall see later, had a detailed discussion of aromatic substitution to the extent it was known at the time of writing.

In contrast, Lucas covered the concepts of Lewis structures in detail, even including how to formulate semipolar bonds, as in sulfuric acid, H_2SO_4. Further, he explained how to determine the so-called 'formal charges' associated with different nuclei in a compound or ion.

Another of the important simple concepts of physical organic chemistry for explaining reactivity to a beginning class is the inductive effect. This follows very naturally after one introduces the idea of polar bonds, such as the carbon–fluorine (C—F) bond. The much smaller electron-attracting power of carbon compared with fluorine in a C—F bond can be rationalized by the Pauling electronegativity scale or, more simply, by the larger nuclear charge of fluorine relative to carbon. However, it must be remembered that comparisons based on these differences may only be correct when comparing atoms in the same row of the periodic table. The concepts of dipole moments fit in well here and Lucas appreciated that.

Differences in electron-attracting powers of atoms are often illustrated by differences in the acid strengths of substituted carboxylic acids of similar structures. The usual comparison is of chloroacetic acid ($ClCH_2CO_2H$) with acetic acid (HCH_2CO_2H), where H and Cl are compared as substituents. We note here that this comparison is not very satisfying pedagogically, because it violates the dictum that the best results will come when the atoms to be compared are in the same row of the periodic table. Regardless, chloroacetic acid turns out to be an 80 times stronger acid than acetic acid. So here the 'inductive effect' operates on the basis that a chloro substituent is likely to be strongly electron attracting like fluorine and more so than either hydrogen or carbon. Both of these latter elements are generally regarded as essentially 'neutral' among the gamut of electron-attracting and electron-donating substituents. The inductive effect of the chloro substituent attracts electrons from the carbon to which it is attached, that carbon then attracts electrons more than usual from the next carbon and so on to the oxygen of the carboxyl ($—CO_2H$), which carries the active hydrogen. Because in acid ionization, that hydrogen is removed as a proton, the inductive effect assists the ionization, by making the key CO_2 group somewhat more electron deficient, less able to hang on to the ionizing proton than it would otherwise. An alternative explanation is a purely electrostatic effect of the $C^{\delta+}—Cl^{\delta-}$ bond transmitted through space. Here, we assume that the positive end of the $C^{\delta+}—Cl^{\delta-}$ dipole is closer to the carboxyl group of chloroacetic acid than the negative end and there will be an electrostatic effect assisting the positive proton to leave by ionization. The question of whether the inductive effect 'travels by train' through the bonds or 'by plane' through space, seems best answered by saying both are important.

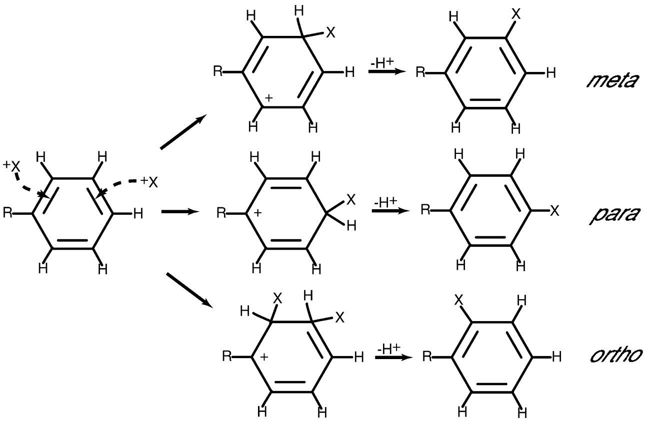
Here, Karrer and Lucas both offer lucid explanations early on. In contrast, the Fiesers mention the difference in acid strengths quite early, but postpone the explanation until rather late in their book. As for amines acting as bases, Karrer is almost silent about what actually occurs, the Fiesers are a bit better and Lucas offers a fairly modern explanation.

A real test of any elementary text in its pursuit of physical organic explanations is in accounting for the orientation, mechanism and reactivities in substitution reactions of benzene derivatives. This has been a classical problem for at least 125 years and the explanations are necessarily complex because no single behavior is observed for all of the types of substitution reactions. However, there is commonality in electrophilic substitutions, such as nitration, sulfonation and some halogenations, because these are featured by attack of an electron-deficient group, symbolized by an entity X^+ seeking to complete its outer shell of electrons by combining with an electron pair supplied by the benzene ring.

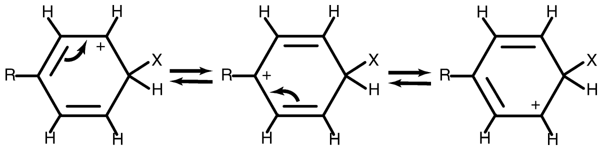
In order to understand in a qualitative way how this leads to understanding orienting effects of the benzene substituents and the observed relative reactivities, it is helpful to write the equations for the reaction of a monosubstituted benzene, where R represents a generic substituent, and the attacking group is the generic, reactive cationic group represented by X^+. The depiction of the substitution processes in Scheme 1 is purposely about the level of sophistication of theory to be expected in the mid-1930s, even at the higher levels of physical organic theory. Of course, a more modern representation would make extensive use of resonance structures or molecular orbitals, but the goal here is to see how our three elementary texts met the challenge of the developing theory in this area of physical organic chemistry.

What we see in Scheme 1, starting from the bottom upwards, is electron-deficient X^+ combining with two of the electrons of a double bond of benzene written in the form of the so-called Kekulé structure with three double bonds. In the lowest equation, X^+ winds up connected to the carbon next to the carbon where R is located and there is a positive charge on the R carbon itself. Loss of H^+ from the carbon where X^+ becomes attached leads to the so-called *ortho* substitution product. The middle equation operates in the same general way, but now X^+ attacks a different double bond to give finally the *para* substitution product. In this case, an extra double-bond shift is required, as shown in Scheme 2, in order to get the plus charge on the ring to be, as it is in the course of *ortho* substitution, with X^+ coming to be connected to the carbon next to R.

The reason for writing the bond shift is that the interaction of R and the positive charge generated by the electron-deficient X^+ attack on the carbon where R is located is the major determinant of the course of substitution. In the upper equation in Scheme 1, it will be seen that X^+ becomes attached to a carbon which results in *meta* substitution, but now in the intermediate stage without the plus charge being on the carbon to which R is attached. This will be true no matter how we shift the double bonds in the way shown.



Scheme 1



Scheme 2. Bond shifts which move the charge around the ring, as might be written in 1930s

To proceed from here is not very complicated, even if it eluded rational explanation for many years. The important thing is that the critical influence of R depends on whether it favors or disfavors having the positive charge on the carbon to which it is attached. If R is a $(CH_3)_3N^+$ group, then having R as a simple positively charged substitutent will favor *meta* substitution and, in addition, will make the overall substitution harder to achieve. This will be because even if X^+ attacks those carbons which will lead to *meta* substitution, the electrostatic repulsions between the charges on R and on the ring in the intermediate will not be favorable compared with R as a neutral or negatively charged substituent.

If R carries an unshared electron pair on the atom immediately attached to the ring, such as F—, HO—, CH_3O— and many other substituents, that electron pair can contribute to the stability of the intermediate resulting from attack of X^+, by forming a bond to the positively charged carbon, for example as $F^+{=}C$, $HO^+{=}C$ and the like. This is normally a very powerful effect favoring what we call *ortho*–para substitution. However, it is subtle, because many R groups of this kind, which have the atom attached to the carbon becoming positive in the intermediate stage, are also ones that more strongly attract electrons than does carbon. A cogent example is fluorine and a carbon—fluorine bond can be written as $F^{\delta-}$–$C^{\delta+}$, where fluorine, because of its larger nuclear charge, pulls the electron pair of the bond closer to itself rather than sharing it equally with the carbon to which it is connected. Such polarizations of bonds were encountered earlier with regard to the operation of the inductive effect in influencing relative acid strengths of carboxylic acids with different substituent groups. Clearly, if R is fluorine, then in aromatic substitution the carbon to which it is attached will be more positively charged than if R was hydrogen. This would, of course, make substitution with X^+ more difficult, and indeed it does. However, the powerful electron-pair sharing effect mentioned above still results in *ortho*–para substitution.

Other possible R groups, such as O_2N—, HO_2C— and NC—, have the atom directly attached to the ring positively polarized as the result of being themselves attached to strongly electron-attracting atoms. Further, those atoms do not carry at least one unshared electron pair so that they cannot act like fluorine and oxygen to use an unshared electron pair to achieve at least partial bond formation and stabilize the intermediate stage.

How do our respective elementary textbooks meet the challenge of the relative rates and preferred positions of substitution on R-substituted benzenes as a function of the nature of R? Here, Karrer simply struck out. He does quote the Pfeiffer and Wizinger's suggestion[10] that chlorine and antimony trichloride could form a complex, which then donates Cl^+ to benzene to form the intermediate stage $ClC_6H_6^+$ and $SbCl_4^-$. However, Karrer does not build on that idea to explain orientation influences or relative rate effects exerted on any of the common substitutent R groups.

Fieser and Fieser, with the advantage of 9 years over Lucas and the likely aid of P. D. Bartlett,[11] produced a thorough and useful presentation of electrophilic substitution. However, there are no ideas in Fieser and Fieser that are absent from the Lucas book. To be sure, the formulas in the two books look very different, but the Fiesers had the advantage of new printing techniques for graphics, which freed them from conventional cold-type representations of organic structures that greatly distorted rings of atoms. It is also true that Lucas correctly reported that many of the standard electron-attracting groups such as $—CO_2R$, $—C{\equiv}N$, and so on, but not $N^+(CH_3)_3$, can tend to withdraw electrons from benzene rings, whereas the Fiesers say that this is only possible with the nitro group. Neither Lucas nor the Fiesers explain the *ortho–para* activating and orienting influence of the methyl group. This was not understood at the time, because the substantial role of methyl groups in stabilizing positive carbon awaited the later definitive studies of Ingold and Hughes on the S_N1 and S_N2 displacement reactions, to say nothing of the concept of hyperconjugation.

For the three reasonably important examples of physical organic concepts, which we might expect to be introduced in the teaching of introductory organic chemistry, it must be seen that Lucas is a clear winner. This is especially so because of his early date of publication and the thoroughness with while he treated the topics.

However, there is more. Lucas covered still other topics not mentioned, or hardly mentioned, by Karrer or the Fieser. Thus, after starting his text with a first chapter providing a four-page general introduction to organic chemistry, Lucas followed this with a much longer chapter on valence, atomic and molecular structure, which broke much new ground for an organic text. There is an excellent discussion of the periodic table, clear descriptions of how to build up Lewis structures, the role of the ionization potentials of the elements, semipolar double bonds, salt formation, hydrogen bonds and the minimum kernel repulsions. The last effect, not much in vogue today, was illustrated by showing how it could help one determine whether $O{=}C{=}O$ is more stable than $O{=}O{=}C$, and $N{=}N{=}O$ more stable than the once-expected $N—O—N$. We can see in this a rather strong Pauling influence.

There were still further subjects that Lucas covered, which clearly Karrer, the Fiesers and other organic chemists of that era were loathe to take up in their teaching, that we now regard as very basic to understanding and using physical organic chemistry. Examples include using bond-energy tables to calculate enthalpies of reaction, the relation between free energy and equilibrium constants, the basics of measuring dielectric constants and distinguishing the difference between polarizability and dipole polarization, and also the idea that water should be thought of as a tetrahedral molecule. Clearly, Lucas was far ahead of his time in introducing physical organic concepts into an elementary organic text and it was essentially 30 years before Morrison and Boyd[12] in 1963 and Roberts and Caserio[13] in 1964 produced elementary organic texts with a strong emphasis on physical organic chemistry, although still lacking some of the subjects touched on by Lucas in his text.

Probably the most important developments in physical organic chemistry subsequent to publication of our three chosen textbooks, which are especially pertinent to elementary courses, were the mechanisms and the relative rates of the S_N1 and S_N2 displacement reactions elucidated by the Ingold–Hughes group. Interestingly, Lucas and his student Saul Winstein contributed much to this development with their studies of neighboring-group effects.

Lucas is probably best known today for three of his PhDs: Winstein, William G. Young and Kenneth Trueblood. He was elected to the National Academy of Sciences in 1957, the only member of the Chemistry Section in the latter half of the 20th century without a PhD degree.

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