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Two textbooks of note: Sidgwick's *Organic Chemistry of Nitrogen* (1910) and Gould's *Mechanism and Structure in Organic Chemistry* (1959)

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ABSTRACT: This whole paper deals with eclipses. The appearance of a book masks the existence of a prior masterpiece. Sometimes, the existence of a book prevents another from even being written. Physical organic chemistry has two such skeletons in its closet. The ghostly presence, Sidgwick's book, contributed to setting up our sub-discipline. The appearance of Gould's useful textbook may well have prevented, this is the second contention, Paul D. Bartlett from writing his own treatise. Copyright © 2005 John Wiley & Sons, Ltd.

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INTRODUCTION

However historians may differ, the surge of a new sub-discipline remains a complex and an opaque process. Contrary to the hypothesis by Lakatos of research programs,[1] a sub-discipline does not spring into existence to fulfil an explicit research program. A sub-discipline gives itself specialized and emblematic organs, such as a periodical for its community of peers and yearly conferences, not only when thriving but especially on the wane, when the initial impetus is gone and when it has run out of gas.

The sub-discipline known as 'physical organic chemistry' may, for the purpose of this paper, be identified with study of mechanisms of organic reactions, of the structure and stereochemistry of organic molecules and of the equilibria in which they are involved. I was active in it throughout my career, from the early 1960s until the turn of the 21st century. I witnessed the officializing of the field after it had enjoyed its heyday.

The periodization[2] which serves as my framework here is a 1910 birth date for physical organic chemistry with the publication of Sidgwick's treatise;[3] a 1934 claim by Ingold for the new territory, complete with new place names; a steep rise in the aftermath of World War II; a steady erosion and a slow decline after the debacle, which occurred in the late 1960s, in public funding of the sub-discipline, following the classical–non classical ion controversy, and the later migration of scientists to more biological topics.[4] Factors explaining the rise of physical organic chemistry in the 1950s were the emphasis on mechanisms of organic reactions when, after the end of World War II, it became once again possible to follow up on the thrust from Henry Eyring (1901–81) and Michael Polanyi (1886–1964),[5] who introduced the notion of a transition state;[6] the lead from the English school of Christopher K. Ingold (1893–1970)[7] and E. D. Hughes,[8] and their use of the kinetic tool; the impact of the first quantum chemical calculations;[9,10] availability of commercial infrared spectrometers;[11] introduction of a novel and powerful spectroscopy, nuclear magnetic resonance;[12–14] the influential and exceptional vitality of a research group headed by Paul D. Bartlett (1907–97) at Harvard[15] (I conjecture a seminal influence by James B. Conant as a mentor to Paul D. Bartlett and Frank Westheimer aiming them towards physical organic chemistry; see W. J. Cromie, *The Harvard Gazette*, 3 October 2002, for F. H. Westheimer's vivid recollection of a comment by Conant redirecting his research); and the sometimes contentious discussions at the Reaction Mechanisms Conferences that were launched in 1946.

I wish to discuss here the influence of two textbooks, one antedating the rise of physical organic chemistry to the first rank by some 30–40 years and the other in its aftermath. The conventional history of physical organic chemistry dates its birth to the 1930s with Ingold[16] or Robinson[17,18] as putative father, in itself a matter of some controversy.

The theme of this paper is eclipses, when the appearance of a book masks the existence of a prior masterpiece. Sometimes, the existence of a book prevents another from even being written. Physical organic chemistry has two such skeletons in its closet. The ghostly presence, Sidgwick's book, contributed to setting up the

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It appeared advisable to take a look at the record, *before* either Ingold or Robinson arrived on the scene. What should that time be? Because physical organic chemistry started its existence before the advent of quantum chemistry and had already set itself apart, it ought to antedate 1930. Were one to stake a claim to the existence of the core notions of physical organic chemistry independently of and prior to electronic theories of molecular structure and reactivity, the ideal time would be the turn of the 20th century.

When did physical organic chemistry start life? I contend here and elsewhere that it came into existence with the publication of Sidgwick's *Organic Chemistry of Nitrogen*.[3] This book issued a clarion call for the new sub-discipline of physical organic chemistry. It outlined its mechanistic research objectives in great detail. It was a pioneering text, at a time when a few other chemists staked out the outlines of the field.

That a new flow of scientific ideas and experiments first takes a subterranean course prior to becoming a mainstream recurs often in science history. The official birth date indeed is recorded a full generation later. As an established sub-discipline within organic chemistry, one may choose as the historical marker the 1940 publication of Louis P. Hammett's influential book,[19] with its title offering furthermore a name for the field.

Textbooks are easily overlooked. Historians seldom turn to them, denigrating them as 'repetitive and uninspired literature'.[20] These scholars are unprepared and even wary of looking at textbooks for, what is much more exceptional, the priming of novel concepts, methodologies and even research programs. And yet! They should remember that the Periodic Table originated in such a textbook, by Mendeleev. Cannizzaro's *Sunto di un Corso di Filosofia Chimica* was also another historical marker. Such examples in themselves ought to focus some attention on chemical teaching manuals. Textbooks provide rich historical evidence on the paradigmatic states of a discipline: subdivisions into areas of expertise; standard problem-solving strategies; standard methodologies and auxiliary techniques; the roster of leaders in the field; . . . .

We are fortunate in the existence of at least two such textbooks in the 1900s, nurturing the rootstock of physical organic chemistry. Julius Berend Cohen's (1859–1935) *Organic Chemistry for Advanced Students*, first published in 1907, went through several editions and was notable for relating mechanisms to kinetics.

THE PRECURSOR

The other book was Sidgwick's *Organic Chemistry of Nitrogen*, which I shall now focus on.[3] This book has had a remarkable career: it has remained in print for over half a century. The first edition was published in 1910, the second edition appeared in 1937 (prepared by Wilson Baker and Thomas Weston Jones Taylor) and was reissued in 1949 and again in 1963 and a third edition revised and rewritten by Ian T. Millar and H. D. Springall appeared in 1967. Sidgwick's contribution was influential, simply because his book (*OCN*) made a deep impression upon organic chemists. Its very circulation makes the point: '25 years after publication copies were selling for four times the original price'.[21]

In arguing that Sidgwick's *OCN* was highly influential, I should mention an obstacle, one familiar to historians of the period: the scientific literature seldom made reference to textbooks, only to primary publications. Education of a scholar saw to it: doctoral students were trained to rely upon original papers exclusively, a rule they heeded afterwards. Accordingly, the proverbial smoking gun proved to be elusive: in spite of all my efforts, I cannot bring forward proof of the direct influence of Sidgwick's textbook upon the nascent physical organic chemistry, in the form of an acknowledgement in print that it served as a standard.

The preface to *OCN* is brief, consisting of only 16 sentences in four paragraphs. It gives *OCN* a dual anchoring, at Oxford and in Germany: Sidgwick pays tribute to the two textbooks of organic chemistry that he holds as authoritative, Victor Meyer (1848–97) and J. Paul Jacobson's (1859–1923) *Lehrbuch* and Victor von Richter's (1841–91) book of the same title.

Even though Sidgwick's preface is short, it constantly evokes the duality between organic chemistry and physical chemistry, but as a border to be bridged. Sidgwick thus pushes for kinetics as a tool to study the mechanism of organic reactions.

A kinetic tool recurrent in *OCN* is the determination of the molecularity for a chemical reaction. Another notion, one of the very foundations of mechanistic chemistry, is that of the slow step: whenever a reaction occurs as a succession of elementary steps, one of them functions as the bottleneck, as the rate-determining step. This concept is also to be found repeatedly in *OCN*, e.g. see Ref. 22.

Mechanistic chemistry is, to a large extent, the study of elusive reaction intermediates. Not only is Sidgwick familiar with the concept, he already adhered as early as 1910 to the modern practice of putting intermediates between brackets (e.g. Ref. 23). Some reaction mechanisms were extremely well understood already by this time. Consider the Beckmann rearrangement: its mechanism is envisaged at several places in the book, and Sidgwick explains how its stereoselectivity can be taken advantage of to assign isomeric structures.[24]

Steric hindrance was already well accepted. The index to *OCN* has no fewer than 11 entries under 'Stereohindrance'. Sidgwick's discussions of stereochemistry are impressive for a book published in 1910. He devotes no fewer than six pages to discussing stereoisomerism in the benzil dioximes.[25]

Another concept already well in place in 1910 is that of ring strain. For instance, diazomethane CH_2N_2 is given the structure of a three-membered ring, to which its high reactivity is ascribed.[26]

Hydrogen and alkyl shifts are covered in Sidgwick's treatise, under the somewhat poetical terminology of 'wanderings'.

Whenever Sidgwick describes a reaction mechanism, in detail the intermediate—apart from becoming enclosed within brackets, for instance a bromamide potassium salt in conversion of amides into amines, under action of alkali and bromine[27]—is placed on the same footing as the other molecules discussed, whether reactants or products. It shares similar structural characteristics as existing molecules. Sometimes, Sidgwick will mention a positively or a negatively charged entity. Such references clearly embody trust in their existence.

Sidgwick's book bridges two periods in time, two shores: on the one hand it is rooted in some of the leading chemistry of the 1890s and 1900s, namely Wilhelm Ostwald's physical chemistry and Emil Fischer's organic chemistry; and, on the other hand, it prefigures physical organic chemistry, as Louis P. Hammett would crystallize its intent when he published his book in 1940.[15]

The impetus of Sidgwick's program, infusing organic chemistry with the tools and concepts from physical chemistry, was so strong as to give it a measure of autonomy and to carry it forward, basically without accommodation of the newer electronic and quantum ideas. These started to be applied about a decade later to organic molecules and reactions by Lapworth and in Robinson's early work.[28]

Prior to the devising of quantum mechanics in the late 1920s, a strong physical chemistry already had deep roots,[29] including within organic chemistry. *OCN* was for several decades a standard reference work. This was the book graduate students starting a career in heterocyclic chemistry were told to peruse at the start of a bibliography. This was the book scientists active in the area of nitrogen-containing organic molecules would consult when writing up their contributions—with the intent of not overlooking work done from the 1880s to the 1900s. This was the book a university lecturer in organic chemistry would turn to during preparation of many a lecture.[30]

This text was highly popular. It was backed-up by formidable documentation. That it carried with it a strong militancy for the nascent physical organic chemistry provided fertile ground. It nurtured a similar spirit among its readers, primarily in Britain and in America. Physical organic chemistry, at least at the start, promised to become an Anglo-Saxon province.[31] (In Germany, while the likes of Hans Meerwein, Walter Huckel or, later, Rolf Huisgen were doing admirable work, they stood somewhat outside the mainstream of organic chemistry.)

OCN has value to the historian of chemistry as strong evidence on two phenomena, each overwhelming in its magnitude, dominance of the field by Germany at the turn of the 20th century, predominance of the kinetic tool.

THE INSTRUCTOR

I will turn now to another textbook. In 1959, Holt, Rinehart and Winston, a commercial New York publisher, came out with a book written by Edwin S. Gould, then at the Stanford Research Institute.[32] Its title was a deliberate inversion of that of the earlier book of Ingold, *Structure and Mechanism in Organic Chemistry*. Gould, by entitling his book *Mechanism and Structure in Organic Chemistry*, emphasized that henceforth mechanistic questions were taking precedence. This textbook ran to 790 pages. Its panorama of physical organic chemistry, even though it was rather slanted and somewhat incomplete, had three parts. The first dealt with molecular structure and with the rudiments of spectroscopy, UV–visible and infrared. The second, which made up the bulk of the book, dealt mainly with reactions involving electrically charged intermediates. The third part, much shorter owing to a combination of unease with its subject matter and a feeling of obligation, dealt with addition and rearrangements proceeding via free radicals.

Gould's preface stated that physical organic chemistry was a mature 30-year-old, which I take as a rhetorical exaggeration on his part. True, one could go back to the end of the 1920s and find memorable ancestors to physical organic chemistry, scientists who indeed were pioneers in the field, such as Gilbert N. Lewis, Arthur Michael, Arthur Lapworth and a few others, especially Louis P. Hammett at the end of the 1930s. Yet they remained isolated and only after the end of World War II did physical organic chemistry soar.

Gould, the author of this monograph, had two somewhat unusual but characteristic features. He lacked direct experience of physical organic chemistry. After he wrote that book, his own work dealt with mechanisms of radical-mediated oxidation in inorganic chemistry. He used the methodology he had earlier been preaching, but applied it to a different part of chemistry. Gould spent most of his career in the chemistry department at Kent State University, in Kent, OH—chiefly remembered today for the National Guard firing upon a demonstration by students against the Vietnam War.

Gould's book, an easy read, a pleasant study guide, clearly and carefully typeset, with an airy format, had behind it sound documentation. As an outsider, the author did a good job overall, making himself familiar with a whole field in spite of its uncertain borders. He had mastered an abundant literature without showing off and is able in this book to distil it in an easily accessible manner.

Moreover, the numerous exercises coming at the end of the chapters, whose answers are not given, are an important feature. They are all stimulating, and some are difficult to solve. A student making the effort of doing them all would benefit greatly and would gain an overview of the field. Often, the background of an exercise would be a question, that had been debated in publications during the 1950s but which would still retain interest at the beginning of the 1960s.

The profusion of exercises was one of the reasons for the success of Gould's book after its appearance. Another reason was that it had cornered the market. Louis Hammett's book, not a textbook and pre-World War II in origin, was no competition. A third reason was that Paul D. Barlett's laboratory had seeded chemistry departments all over the USA with well-trained, gifted, physical organic chemists. As insiders, they understood the material far better than Gould did. For them, it was an ideal situation. They could assign Gould as reading and for the exercises, and they could lecture from their own material. They could dispense with the effort of writing up their lecture notes, while they could afford to be critical and even highly critical of Gould's.

Gould's book is most valuable to the historian of chemistry for its documenting of everyday life in the laboratory during the 1950s. It makes us see graduate students measuring reaction rates, by conductimetry for instance, in carefully thermostated reaction vessels; determining the magnitude of an electric dipole moment; using calorimetry to determine the heat of a reaction, and then pitting it against a value calculated from bond energies; scribbling Lewis structures on the back of an envelope, to estimate a bond length or an electric charge . . . .

COMPARING TWO COMPILERS

Now to a parallel, of sorts, between these two books, Sidgwick's *Organic Chemistry of Nitrogen* and Gould's *Mechanism and Structure in Organic Chemistry*. They had a joint fate. Neither became a landmark for physical organic chemistry as a sub-discipline. In the 1960s, when physical organic chemistry was at the acme of its popularity and appeal among chemists, Sidgwick's book remained totally forgotten among physical organic chemists. Synthetic organic chemists, natural products chemists, specialists of alkaloids in particular and pharmaceutical chemists continued to give it their allegiance.

There are two explanations for the neglect by physical organic chemists. It was a book which was ahead of its time, the work of a pioneer. Moreover, Sidgwick went on to a different field where he made his name. One might say that he defected to the enemy, he became a militant of the electronic theory of chemistry. His second book, *The Electronic Theory of Valency*,[33] made him famous. When he was awarded the Medal of the Royal Society in 1937, it was stated that this book had more influence on British science than any other in his generation. The success of the second book eclipsed almost totally the merits of the first. Furthermore, readers of the second book, as they became convinced of the pertinence and cogency of the electronic theory, took it for granted that Sidgwick's earlier approach had been superseded.

And what about Gould's book and its reception? It stood in the shadow of a non-existent book. It would have been natural, in the late 1950s or early 1960s, for Paul D. Bartlett to write the authoritative masterpiece for which he was uniquely qualified. He did not. Steven J. Weininger may tell us the reasons when he publishes the book on Professor Bartlett on which he is currently working.

An early reader of the present paper noted that Bartlett himself immediately adopted Gould's text for his own course and never seemed to criticize it explicitly (at least from 1961 to 1965, when he attended his lectures)—a courteous behavior totally in character for Paul D. Bartlett. This former Bartlett student further reminisced, 'of course he did not follow Gould in his lectures'.

In any case, Gould's book pre-empted this particular niche of the market. Only a decade later would Bartlett publish a book, an annotated collection of reprints from the classical–non classical carbonium ion controversy, that reflected his eminent skills as an analytical thinker, a gifted writer and a fully informed scholar.[34] By contrast, Gould's book lacked authority. Its writer did not have the qualifications that would otherwise have put his volume in the category of Hammett's or of *The Nature of the Chemical Bond*.

Comparison of those two books, Sidgwick's and Gould's, across half a century has value for the historian. Even though neither book advocates a methodology, as Hammett's did with linear free energy relationships, Streitwieser's manual[35] or Coulson's[36] with molecular-orbital calculations, Dewar's with perturbational MO theory,[37] they give witness to the anticipated (by Sidgwick) rise of a new field. They also document its migration across the Atlantic. Their continuities include the importance of the molecular formula as a heuristic tool and the measurement of reaction rates, whereas the discontinuities have to do predominantly with the notion of a transition state as distinct from an intermediate, and potential energy diagrams along the reaction coordinate.

Gould's book indeed reflected a core weakness of physical organic chemistry. If one excepts a few scientists such as John D. Roberts and Saul Winstein, its blind spot as a sub-discipline was the absence of quantum chemical calculations. Gould, in the mainstream of organic chemistry in the 1950s, contented himself with qualitative use of valence-bond theory. His book could have been the opportunity for the introduction of molecular orbital theory, if only at the level of the Hückel approximation. Indeed, the appearance of contemporary introductions of molecular orbital theory aimed at organic chemists—I am alluding to books such as those by Andrew S. Streitwieser[38] and John D. Roberts[39,40]—makes his lapse even more glaring to our *post hoc* perception.

Gould had an excuse, though. He shared this blind spot with physical organic chemists as a group, as a community. At the end of the 1960s, this particular deficiency would lead to the implosion of physical organic chemistry. For one thing, the classical–non classical ion controversy pitted people trained in physical organic chemistry as a heuristic, against scientists who had an interest in and an understanding of quantum theory. One may draw a parallel with the controversy about stereochemistry between van't Hoff and conservative chemists in the mid-1870s, or about atomic theory among French chemists in the 1880s and 1890s. In such episodes, a group of conservative chemists resisted innovation in the name of empiricism and to combat what they held to be only idle speculation.

To return to the impact of quantum chemistry and calculations on organic chemistry, the advent of the Woodward–Hoffmann rules for electrocyclic reactions proved that quantum theory was successful, that it was predictive and that the toolbox of traditional physical organic chemistry was by then, around 1965, hopelessly dated. This was when the rules of the game started to change.

CONCLUSIONS

What do we learn from these two books, their times of appearance, their contents, their reception, about the constitution of a scientific sub-discipline? Any field of knowledge is highly vulnerable. The troubles of physical organic chemistry at the end of the 1960s, one year towering so high, a couple of years later fallen from its pedestal and replaced by synthetic organic chemistry, is an eloquent testimony to the fickleness of scientists as they follow the grant money.

Not only are they fickle, their collective memory is weak and selective. Any field of science oblivious of its historical roots is set for disaster, sooner or later. The reason is that concepts are not God-given. Rather than appearing from nowhere in the big blue yonder, they are set on foundations and they are constructed, individually and collectively, in a gradual combination of accretion and demotion.

If there is a lesson to be drawn from the two highly different books on which I have been commenting, it is that in the short run opportunism pays. Not only is it rewarded with success but, as the saying goes, bad currency drives out the good. At its zenith, physical organic chemistry was beset with a number of mediocre manuals. Moreover, publication by Gould of his very good book may well have prevented the publication of a Sidgwick-like treatise, which is what Paul D. Bartlett could most certainly have provided.

A lesson from Sidgwick's book is how important it is, how determining of the future it is, to review a field critically and to sum it up. Owing to his efforts, and to those of a few of his contemporaries, the field of physical organic chemistry was prepared already in the 1930s, it needed only to be harvested. Sidgwick's accomplishments are summarized in one word, scholarship.

The lesson from Gould's book is similar. When we know from hindsight that physical organic chemistry would not reach unharmed the end of the 1960s, can we put some of the blame on Gould's book? Only to the extent that it was loyal to an already outdated and unduly restrictive view of the field, which excluded chemical physics and quantum chemical calculations.

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