THOMAS MARTIN LOWRY AND THE MIXED MULTIPLE BOND

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Thomas Martin Lowry (1874-1936) is a name familiar to all chemists primarily for his theory of acids and bases. He also discovered, named and elucidated the mechanism of mutorotation as well as being a pioneer in the study of optical rotatory dispersion. Deeply interested in the history of chemistry he published a widely used text *Historical Introduction to Chemistry* which went through three editions (1). Trained originally as an organic chemist his interest gravitated to physical chemistry. Lowry was one of the pioneers in the developing hybrid discipline of physical organic chemistry. With his student William Alec Waters (1903-1985) he produced in 1935 the first book in this area, *Physical Aspects of Organic Chemistry*. Others by Herbert Ben Watson (1894-1975), Louis B. Hammett (1894-1987), and Gerald Branch (1896-1954) and Melvin Calvin (1911-1997) followed in 1937, 1940, and 1941, respectively. One of Lowry's most significant contributions to physical organic chemistry was his early advocacy of the use of Lewis-Langmuir theory in the interpretation of organic reactions (2). This paper will review and assess the significance of his role in this formative period of physical organic chemistry.

Lowry was born October 26, 1874, in Bradford, England. His father was a Methodist minister as had been generations of Lowrys before. His life was dominated by a pious devotion to his religion; and his biographers Allsop and Waters (3) state that his scientific career was always guided by his devotion to his religion. A clue as to why Lowry would gravitate to physical organic chemistry comes from his academic training. In 1893 he entered the Central Technical College of the City and Guilds Institute in London to embark upon his chemical training. There he came under the influence of Henry Edward Armstrong (1848-1937), an iconoclastic teacher and researcher. Armstrong, an organic chemist, is best remembered for his centric formula for benzene; but he also did significant work on

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the stereochemistry of natural products and chemical crystallography. Armstrong was one of the first to cross the traditional boundaries between physical and organic chemistry. On his graduation in 1896 Lowry became Armstrong's assistant and remained so until 1913. During this period he obtained his D.Sc. degree (1899) and

taught himself physical chemistry (4). He held a series of teaching positions in this period which culminated in 1912 in a Lectureship in Chemistry at the Guy's Hospital Medical School. His reputation as an innovative researcher led to his election as a Fellow of the Royal Society (1914) and his promotion to the rank of Professor. In 1920 the newly created physical chemistry chair at Cambridge was offered to Lowry, and he spent the rest of his life at Cambridge. In summarizing his work his biographers have written (3):

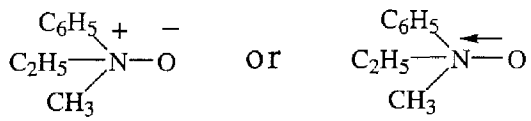
> Lowry gained scientific eminence as a physical chemist, and for the last twenty years of his life held one of the senior professorships of physical chemistry in England; but at the outset of his scientific career he was an experimental organic chemist busily engaged in elementary teaching. His early work in both these directions deeply influenced his outlook throughout his whole life, for his approach to physical chemistry was always that of the experimenter rather than that of the theorist, possibly because his mathematical knowledge was limited, though in this direction he was enthusiastic in stimulating the efforts of collaborators.

Lowry became one of the most enthusiastic advocates of the Lewis-Langmuir theory. His original contributions to the field were to occur in the period only from 1923-1925, but his impact was to be long lasting.

The characterization of the electron by J. J. Thomson of Cambridge University (1856-1940) in 1897, as a fundamental sub-atomic particle in 1897 was followed by initial speculations on the role of the electron in chemical bonding. Thomson's first paper on the subject appeared in 1904, but it seemed to have had little impact on the British chemical community. Thomson's conception of bonding was a modern revival of Berzelius's dualism. His ideas were enthusiastically adopted by a group of American organic chemists who attempted to explain chemical properties as well as reaction mechanism based upon the Thomson model. This American effort was doomed to failure and would in turn in the United States retard the adoption of the newer ideas developed by the American chemists G. N. Lewis (1875-1946 in 1916 and Irving Langmuir (1881-1957) in 1919. American organic chemists with a few exceptions such as James B. Conant (1893-1978) and Howard Lucas (1885-1963), did not get involved in the application of Lewis-Langmuir theory. British chemists who had not been bruised by the battles fought earlier in America were far more receptive to new ideas.

Lowry's interest in the Lewis-Langmuir theory began sometime after 1921. Langmuir had spoken at length on his octet theory at the 1921 British Association for the Advancement of Science meeting in Edinburgh. J.J. Thomson had over the years modified his views from a strictly electropolar approach to accommodate electron sharing and covalence. These ideas were to appear in papers in 1907 (5), 1914 (6), and in 1921 (7). Thomson could not envision bonding as a gradation of the extent of electron pair sharing. To him polar and nonpolar bonds were distinctly different. Lowry's first contribution to the electronic theory occurred in 1922, when he read a paper before the Faraday Society on November 20, entitled "The Electronic Theory of Valency. Part I. Intramolecular Ionization" (8). This paper appeared in the February, 1923, issue of the *Transactions of the Faraday Society*. Most of the paper is devoted to examples from inorganic chemistry, with a few examples from organic chemistry. The key premise is that "a condition of intramolecular ionization exists in a large number of compounds, where nothing of the sort has been suspected previously" (8). With respect to amine oxides Lowry writes (8):

> It therefore really looks as if the obvious method of representing this compound; as one in which the oxygen is linked to the nitrogen by a mixed double bond, including one covalency and one electrovalency, thus may perhaps be novel.



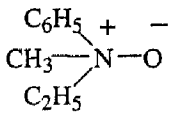
Lowry suggested this structure in response to that postulated by-Langmuir for amine oxides (8):

> Langmuir states definitely that in this compound the nitrogen is quadricovalent, and it is entirely unnecessary to assume a quinquivalent nitrogen. It is difficult to believe that Langmuir can have overlooked the fact that in this system the nitrogen atom is positively charged (and thereby converted into a kation), whilst the oxygen is negatively charged, and so converted into a bound anion.

On November 27, 1922, shortly after the Faraday Society meeting paper, Lowry wrote to G.N. Lewis. The letter was an acknowledgment by Lowry of reprints of recent work by Lewis. Plans to confirm a visit by Lewis to England in the summer of 1923 were also included. The post-script is the most important part of the letter and is reproduced below (9):

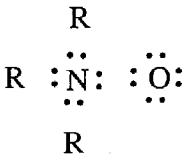
> I should be interested to know your opinion in reference to Langmuir's electronic formula for N_2. My

> own view is that it is an anomaly for which I cannot find any sufficient justification; and I certainly prefer the interpretation which you gave in your 1916 paper. I have also been interested myself in the discovery of electric charges on the atoms in the electronic formulae of quite a lot of simple compounds, e.g. methylethylaniline oxide.



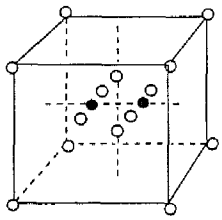
> All this appears very obvious, but I cannot find that it has been published before, and it certainly leads to some interesting results both in inorganic and in organic chemistry.

Lewis in his own hand wrote the following structure:



Neville Sidgwick (1873-1952) in 1927 was to describe the bonding in amine oxides as a coordinate covalent (or dative) bond. He writes "this is the semi-polar link of Lowry and others which is written $\overset{+}{A}—\overset{-}{B}$ or $A—B$ (10). Lowry never used the terms "coordinate" or "dative" in his writings.

Langmuir had presented a rather odd structure for the nitrogen molecule in his 1919 paper "Arrangement of Electrons in Atoms and Molecules" (11). Seeking to use the Lewis cubical atom, he presented the structure shown below, in which an electron pair is embedded in the octet that surrounds the two nitrogen atoms in order to accommodate the 10 electrons of the two nitrogen atoms. This in essence was a quadruple bond.



Lewis had realized in 1916, that using the cubical atom concept would make it impossible to represent triple bonds or to show the concept of free rotation, a basic tenet of stereochemistry. He states (12):

> On the other hand, the group of eight electrons in which the pairs are symmetrically placed about the center gives identically the model of the tetrahedral carbon atom which has been of such single utility throughout the whole of organic chemistry.

Thus in N_2 there are three bonds, the maximum number Lewis believed could exist, and he represented the molecule as :N:::N: .

Langmuir seemed never to have mastered the difficulties and problems of using the cubical atom in more complex situations in organic chemistry. His 1919 paper is replete with attempts to use the cubical atom instead of dot formulas to represent structures. Lowry was adept enough at this point to see that by using the electron-pair and octet concepts it was not possible to write a formula for amine oxides without formal positive and negative charges. Lewis never adopted Lowry's suggestion, although he comments in his 1923 monograph on "Valence and the Structure of Atoms and Molecules" as follows (13):

> Presumably such a substance as amine oxide is therefore considerably polarized.

Lewis was probably loathe to use signs because it would look like a revival of the electropolar theories he had so thoroughly discredited. Lowry's representation of amine oxides is, of course, used in all textbooks today.

Over the next few months Lowry extended his ideas on the electronic interpretation of multiple bonds in organic compounds. These were to become his most significant contribution. In a letter dated January 3, 1923, to Lewis, Lowry set forth his ideas on the nature of these bonds. Before giving his rationalization for multiple bonds he returned to the problem of formal charges in amine oxides (14).

> If so, you will see that the electric charges that I have postulated are arrived at by dividing equally between the two atoms the duplet which constitutes the bond been the oxygen and nitrogen in the amine oxides. The mere balancing of the electrons against the nuclear charges then gives evidence of an excess or a deficiency of electrons on the individual atoms.

With regard to multiple bonds he states the following (14):

> As regards the formula for N_2, I am inclining more and more towards the view that unsaturation nearly always means the presence of electrovalencies, usually in the form of a 'mixed' double or triple bond containing at least one valency of each type. Thus the unsaturation of ethylene and acetylene can be ascribed to the association of one electrovalency with

> one or two covalencies, respectively, as in $\overset{+}{C}H_2-\overset{-}{C}H_2$ and $\overset{+}{C}H=\overset{-}{C}H_2$. On the other hand, the saturated character of O_2 and N_2 would be ascribed to the presence of double and triple covalencies.

Lewis in his structure for ethylene which he presented in 1916 has two formulas: H:C::C:H and H:C:C:H. In a cryptic note Lewis writes, "I shall postpone a discussion of the important bearing of such formulae upon the problem of the conjugate bond" (15). Unfortunately this seemed to have been a permanent postponement! Certainly Lowry's conception was different from Lewis' as to the nature of the double bond.

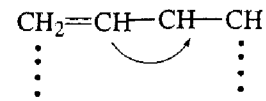
Who were some of the persons and the ideas that influenced Lowry to produce his "novel" conception of unsaturation? Certainly J.J. Thomson was one. On January 6,1923, Lowry wrote to Thomson the following concerning a lecture Thomson had recently given in which he discussed the electronic nature of unsaturation and conjugation. Thomson presented a model in which the reactivity of the double bond was the result of dividing the bond equally, in essence producing a septet at each carbon atom. Thomson used the cubic model of Lewis to show how this happened (16).

> I was interested to hear your references to conjugated compounds in your lecture on Thursday morning. There are two possibilities as regards the opening of the hinge between two doubly bound carbon atoms. I think your scheme involved the formation of two septets: but I have been following up the alternative hypothesis that one carbon atom takes both electrons giving rise to an octet and a sextet. This makes the alternate atoms positively and negatively charged so that the formula for butadiene becomes
>
> $\overset{+}{C}H_2-\overset{-}{C}H-\overset{+}{C}H-\overset{-}{C}H_2$

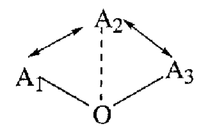
J. J. Thomson in reply seemed to accept this idea of Lowry as a possible electronic interpretation of the double bond. This is one of the earliest known examples of what became known as the electromeric effect.

Another important influence was the work of Johannes Thiele (1865-1918). Thiele, as distinct from most German organic chemists of his time, was very much interested in the theory of organic reactions. In 1899 he proposed his theory of partial valencies to explain the phenomenon of conjugation and the apparently anomalous chemistry that results. All atoms had inherently some residual affinity for reaction, according to Thiele. Thus in butadiene the inner two carbon bonds became saturated from the residual affinity on C_2 and C_3, leaving partial valencies on the terminal carbons to produce enhanced reactivity

$CH_2=CH-CH=CH_2$



Hugo Kaufmann (1870-?) in a 1908 paper in the *Physikalische Zeitshrift* (17) interpreted Thiele's theory in terms of the electron theory of valence. He argued that valence was divisible and that the lines of force associated with the electron were divided among three atoms. Thus although three atoms A_1, A_2 and A_3 shared a pair of electrons to form a bond, the valence was distributed over three atoms.



(after Stranges, Ref. 2)

Kauffmann's electronic structures bear strong similarities to those of Thiele. His dashed and dotted lines were equivalent to an electron bond in the former and a partial bond in the latter. To explain the reactivity of unsaturated molecules which was not obvious from the structural formula, Kaufmann proposed that in these systems there were more numerous lines of force more widely extended out in space, thus creating enhanced reactivity. Kauffmann's interpretation did not seem in retrospect to elicit much support or use in the chemical community. It was only a small step for Lowry to translate the formula of butadiene of Thiele to the one he proposed to J. J. Thomson.

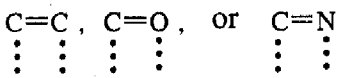
In a paper received on March 14, 1923, by the Chemical Society and appearing in the April, 1923 issue, Lowry expanded on his view of the double bond in organic chemistry. The paper opens as follows (18):

> The object of this paper is to suggest that, whilst a single bond may be either a covalency or an electrovalency, a double bond in organic chemistry usually reacts as if it contained one covalency and one electrovalency. Acetaldehyde is therefore written as
>
> $CH_3-\overset{+}{C}H-\overset{-}{O}$ and ethylene as $\overset{+}{C}H_2-\overset{-}{C}H_2$
>
> Bonds of this character are described as mixed double bonds.... Each (-) sign then indicates an excess of one planetary electron above the net nuclear charge, and each (+) sign a deficit of one electron.

According to Lowry an electrovalency enhanced the reactivity of any substance, regardless of the elements.

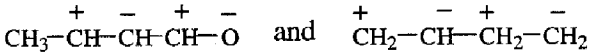
The enhanced reactivity of double bonds versus single bonds is the result of the mixed bond in the former. Thiele's ideas of partial valencies are easily translatable on an electronic basis by using the mixed bond:

The superposition of an electrovalency on a covalency therefore provides a convenient explanation of the subdivision of affinity, which Thiele indicated by dotted lines in unsaturated groups such as



These may be written as C=C, C=O, and C=N, where oxygen and nitrogen are usually negative relatively to carbon, and oxygen relatively to nitrogen.

Perhaps the most important insight Lowry had in this paper deals with conjugated compounds. These were the subject of much theoretical speculation in the 1920's by the British chemists Arthur Lapworth (1872-1941), Robert Robinson (1886-1974), and Christopher Ingold (1893-1970) (18):

> Crotonaldehyde and butadiene are formulated as
>
> 
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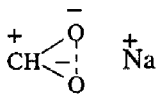
> It will be seen that under this scheme the distinction between single and double bonds in a conjugated system disappears completely. There is therefore no longer any need to postulate a wandering of the double bond when butadiene is brominated, since the central double bond is already in position.

From our present perspective we could read into this statement the concepts of mesomerism as well as electromerism. These would become part of the system of tautomeric effects involving polarization and polarizability that Ingold was to develop beginning in 1926. Similarly Lowry's explanation of the structure of the ions of carboxylic acids is very much as used today. For example in sodium formate "The distinction between the single and double bonds disappears, just as in the case of conjugation" (18).



Other applications were made by Lowry by using the mixed bond which were to lead to controversy. Lowry firmly believed that there were formal charges in the systems he discussed and he compared his ± sign as the same as that used in compounds such as sodium chloride. Statements of this kind were bound to cause Lowry problems. Samuel Sugden (1892-1950) of Birkbeck College, London, almost immediately called Lowry to task in a short note received by the *Journal of the Chemical Society* on June 21, 1923, entitled "Electron Valency Theory and Stereochemistry" (19). Sugden points out that Lowry's conception of the double bond would lead to free rotation and the lack of geometrical isomerism. Lowry quickly replied on July 10, 1923 and this followed Sugden's note (20):

> In reading my paper on 'The Polarity of Double Bonds' I laid considerable stress on the fact that the formulae there set out represented the reactions of the various compounds rather than the resting-states of the molecules. The metaphor used was that these formulae represented 'the dog standing up and barking' and that he might assume a very different attitude 'when curled up and at rest.'

In some cases such as the amine oxides there is no distinction between the ground and activated states. Lowry implied that the mixed double bond is only operative in the activated state and until a molecule is activated ordinary considerations will apply. The use of the ± signs by Lowry and his qualification in this reply to Sugden created confusion. Lowry recalled in a letter dated May 9, 1935, to William Albert Noyes (1857-1941) of the University of Illinois (21):

> These proposals met with such fierce opposition that for some years it was unwise to write a + or - sign on the blackboard of the Chemical Society for fear of open ridicule by Ingold and others.

In the midst of writing these papers Lowry was organizing what was to become a landmark conference in the application of electronic theory to organic chemistry. A two-day meeting on July 13-14, 1923 was held at Cambridge under the auspices of The Faraday Society. The symposium entitled "The Electronic Theory of Valency" consisted of three sessions. G. N. Lewis gave the introductory address entitled "Valence and the Electron" (22). Among those attending were J.J. Thomson, William Bragg, William Albert Noyes, Arthur Lapworth, Robert Robinson, R. G. W. Norrish, Jocelyn Thorpe, Ian Heilbron, George Norman Burkhardt, and Bernard Flurscheim. Many of these were already the leaders or soon to be leaders in British organic chemistry. In particular Lapworth and Robinson had been very active in the area under discussion.

Lowry presented the introductory address to the second part of the symposium which dealt with applications to organic chemistry. Lowry defended his concept of the mixed bond as a convenient way to explain the reactions but probably not the structure of a variety of unsaturated compounds and conjugated systems (23):

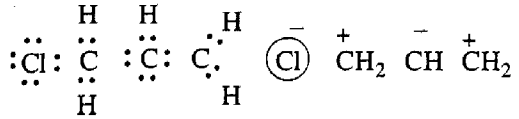
> For good or evil, it possesses one feature which distinguishes it from some other electronic theories. Thus, although it was definitely suggested by and is firmly based upon the electronic theory of valency, it is capable of being expressed by familiar symbols, the meaning of which is readily grasped by all chemists. It therefore presents a specially easy target for criticism, of which it has received at least a full share. These criticisms appear to me to have done no damage to the target; and, whilst questions may be raised as to the conditions under which the structure that I have discussed is developed, I am confident that the mixed double bond represents a real alternative to the well-recognised double covalency.

Lowry's original contributions to the symposium were two papers: "Intramolecular Ionisation in Organic Compounds" (24) and "The Transmission of Chemical Affinity By Single Bonds" (25). In the former he presented evidence showing that a polar catalyst is required to cause ethylene to react with bromine which was in line with the idea of an activated double bond being a mixed bond. In the latter paper he addressed the question of the types of valency and methods for the transmission of these. All the papers presented at the symposium, including the discussion that ensued, were printed in *The Transactions of The Faraday Society*.

In many of the papers and the discussions which followed exception was taken to Lowry's interpretation of the double bond. Lowry proposed that the mixed bond has to be thought of in the context of the problem of structure and reactivity. In the ground state the electron pair will be equally shared, but during the reactive phase there may be an ionization of the bond to produce the mixed bond. Lowry stated (26):

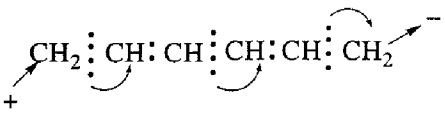
> At the end of the discussion I am still convinced that the mixed double bond, in which one of the links has been completely ionised, really exists and plays an important part in many branches of organic chemistry.

At this point it would be instructive to compare the ideas of Lowry with those of Robinson, who was also initially influenced by Thiele's residual affinities concept as well as by the alternating polarities of his colleague Arthur Lapworth. In a 1922 paper written in collaboration with William O. Kermack (1898-1970) entitled "An Explanation of the Property of Induced Polarity of Atoms and an Interpretation of the Theory of Partial Valencies on an Electronic Basis" (27), the attempt was made to translate Thiele's and Lapworth's ideas by using Lewis-Langmuir theory. In discussing the reactions of conjugated systems Kermack and Robinson produced the concept of the mobility of the octet in response to some internal or external agent. This would lead to an alternation of polarities represented by + and - signs. These signs were not to be taken in the same light as Lowry's. For example, allyl chloride could be represented in the following way (27):



Here the stable octet surrounding the chlorine atom produces an unstable system about the adjacent carbon atom, and therefore the formation of a stable system in the CH group is facilitated by the aid of two of the electrons held in common with the second unsaturated carbon atom. It is accordingly quite natural that the product of the addition of H Br is trimethylene chlorobromide $Cl–CH_2–CH_2-CH_2Br$.

In discussing chemistry of conjugated systems such as hexatriene, Kermack and Robinson chose the following representation by incorporating the curved arrow:



Robinson's view of multiple bonds was far more general than that of Lowry. He viewed the enhanced reactivity in terms of the mobility of the octet, not an implicit electrovalency. For this and other reasons Robinson proved to be far more successful. Colin Russell, in his classic study of valence, best summarizes the contributions made by Robinson and Lowry; (28):

> Thus the covalent bond was no longer conceived as a rigid entity with two electrons symmetrically shared between the atoms. A molecule was subject to strains and stresses unimagined twenty years previously, and the valency bonds were more dynamic than static, responding to the demand of a reagent to assist a reaction.

Lowry's involvement with further developments in the electronic theory of valency after 1923 was to be peripheral in nature. In a brief paper published in the *Philosophical Magazine* in 1924 entitled "The Origin of Acidity," he stated his definition of acidity as follows (29):

> An acid may be defined as a hydride from which a proton can be detached, e.g. on dissolution in an ionizing solvent, on electrolysis, or by displacement by a metallic ion.

To explain the order of acidity in the series CH_4, NH_3, OH_2, and HF, Lowry adopted ideas from the Bohr model

of the atom. He invoked the idea that the electrons in a molecule are in what he called looped orbits which execute a figure eight between the nuclei in a molecule. In molecules like methane the loops are for removed from the nucleus. There is little probability of repulsion between electrons in these orbits and thus the chances of losing a hydrogen as a proton are minimal. However in hydrogen fluoride for example (29):

> ...the much smaller loops in which the proton of hydrogen fluoride is held must bring it nearer to the nucleus, and may easily bring it so near that it can be driven outside the loop by the strong repulsion to which it is subjected.

In order to explain the transmission of acidity through a carbon chain, such as in halogenated aliphatic carboxylic acids, the suggestion was made by Lowry that atoms like chlorine tend to change the size of the orbits of the electrons they share with other atoms. Thus in a dynamic way this will cause alterations of properties along a chain whose effect will diminish with distance. Groups that enhance basicity will function in the opposite manner. These novel suggestions indicated the way that Lowry, essentially a product of the nineteenth century, so eagerly embraced new ideas.

Although few chemists were to adopt Lowry's concepts of the mixed or semi-polar bond, his faith in these ideas persisted through 1925. For example, in a discussion at the British Association meeting in Southhampton on September 1, 1925, devoted to "The Alternating Effect in Carbon Chains," Lowry offered what he believed was new evidence for his views (30). Sugden had developed a concept in 1924 (31) called the parachor, a measurement of molecular volume by use of surface tension, density measurements of liquid and vapor phases, and molecular weight [P = M 1/4]. Parachor measurements, according to Lowry, were inconsistent with a nonpolar double bond in such systems as nitro, for example. There must be a nonpolar as well as a polar bond, and a group such as the sulfoxide contains only a polar bond.

In 1925 Phillips reported resolution of the compound ethyl toluenesulfinate into a pair of enantiomers. Lowry saw in this report conclusive evidence for the semipolar bond. As Lowry stated, the S=O bond in these compounds must be semipolar since only when represented by the symbol

$$\overset{+}{\text{S}}\!-\!\overset{-}{\text{O}}$$

could the existence of optical activity be possible. Sugden's measurement of the parachor for this compound also offered additional evidence for the semipolar bond. Sugden's parachor was succeeded by more sophisticated physical measurements; indeed, the optical activity of the sulfinates is based on an entirely different explanation. Nevertheless, Lowry certainly tried to use all the experimental evidence of his day to validate his ideas.

To his credit, Lowry was one of the few university lecturers in Britain in the 1920's who dealt with the electronic theories of valency in his courses. His student W. A. Waters reminisced (32):

> At Cambridge in my time (1923) he gave a wide set of lectures on recent theories of atomic and molecular structure, following Bohr, Lewis, and Langmuir, choosing his examples as much from organic as from inorganic chemistry, and giving the early spectroscopic evidence that led to the quantum theory of atomic and molecular orbitals.

Although Lowry's contributions to the electronic theory of valency may be considered only as a footnote to the work of others, through his prestige he certainly was influential in setting the state for the remarkable developments that occurred in Britain in the era between 1919-1939.

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Letter to the Editor

In Nos 15/16 of the *Bulletin*, the paper by Pierre Laszlo "Georges Darzens (1857-1954): Inventor and Iconoclast" pages 59-64 was marred by a most unfortunate editorial addition: at no time in its history was the Ecole polytechnique headed by a German general! Even though this horrible mistake was corrected on the proofs, Mr. Murphy or the vengeful Gods left it for all to see!
– Pierre Laszlo