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Photo on opposite page courtesy of the AIP Emilio Segrè Visual Archives, Margrethe Bohr Collection; sodium sketch adapted from reference 7 .

Published in a series of three papers in the summer and fall of 1913, Niels Bohr's seminal atomic theory ¹ revolutionized physicists' conception of matter; to this day it is presented in high school and undergraduate-level textbooks. However, the theory is usually understood to pertain merely to one-electron atoms, with which it scored its most spectacular successes. From a historical point of view, that is a gross misconception, for Bohr originally thought of his brainchild as a much more ambitious theory that would lead to a new understanding of the constitution of all matter, whether the physicist's atoms or the chemist's molecules. After all, the very title of his publication, "On the constitution of atoms and molecules," indicates that it was addressed as much to chemists as to his colleagues in physics. Whereas Bohr focused on the hydrogen atom in the first part of his work, he devoted the second part mostly to more complex atoms and the third to the structure of molecules. In that 1913 trilogy, considerations and results of a chemical nature played a significant role often overlooked today. Moreover, his theory had important chemical consequences, in particular regarding the periodic arrangement of the elements.

The often contentious relationship between the two sister sciences, physics and chemistry, is a theme that can be followed through much of the history of science. In his post-Newtonian *Metaphysical Foundations of Natural Science*, dating from 1786, philosopher Immanuel Kant argued that chemistry could never be a genuine science such as the much-admired mechanical physics, because its subject matter was intractable to mathematization and did not follow by necessity from the laws of nature. Later chemists continued to look forward to a "Newton of chemistry" who would base their science deductively on the higher principles of physics. Was Bohr's theory perhaps the answer to the call? Arnold Sommerfeld in Munich thought that it might well be and that chemistry was now on its way to being reduced to physics. In the UK, Oliver

Lodge saw Bohr's atomic theory as a new chapter in the history of the relationship between the two sciences. It looks, he said, "as if it were going to do for Chemistry what Newton did for the Solar System."² But that is not what happened. In spite of all its promise, Bohr's semiclassical atomic theory did not succeed in explaining chemistry on a purely physical basis.

When the 18-year-old Bohr enrolled at the University of Copenhagen in 1903 to study physics, he was to receive a solid, all-around education in mathematics, astronomy, and chemistry. In 1905 he met the six-years-older Niels Bjerrum, who taught an experimental course in inorganic analytical chemistry. As Bjerrum recollected, Bohr was an eager but somewhat clumsy student of chemistry who caused the laboratory quite an expense in broken glassware.³ Some years later Bjerrum would begin groundbreaking studies in the application of quantum theory to rotating and vibrating molecules. He was also one of the first chemists to refer to Bohr's atomic theory. In a 1917 Danish chemistry textbook, he made use of the new Bohr–Sommerfeld model of atomic structure, particularly its definition of a chemical element in terms of nuclear charge Z rather than atomic weight. For his part, Bohr was acquainted with Bjerrum's work in molecular spectroscopy, to which he referred in the third part of his trilogy. Bohr and Bjerrum became lifelong friends, sharing not only scientific interests but also a sailing boat named Chita.

After having settled in as a professor of physics at the University of Copenhagen, and especially after the founding of the university's Institute for Theoretical Physics (now called the Niels Bohr Institute) in 1921, Bohr was in regular contact with Danish chemists Jens Christiansen and Johannes Brønsted. Christiansen, who worked for a period at Bohr's institute, was impressed by Bohr's great flair for chemistry, which he in part ascribed to a natural faculty for chemical problems and in part to Bohr's contact with chemists.

Foremost among those who inspired and interacted with Bohr was Hungarian physical chemist George de Hevesy, with whom Bohr established a lasting friendship. Hevesy, who would receive the 1943 Nobel Prize in Chemistry for his work on radioactive tracers, was receptive to Bohr's early ideas about atomic structure and the origin of radioactivity. In a letter to Hevesy dated 7 February 1913, Bohr outlined some of the chemical content of his as-yet unfinished theory, including "a very suggestive indication of an understanding of the periodic system of the elements" and an explanation of how atoms combine to form molecules. Generally, the ambitious Bohr envisaged "a detailed understanding of what we may call the 'chemical and physical' properties of matter."⁴

In addition to being awarded the 1922 Nobel Prize in Physics, Bohr was nominated twice for the Nobel Prize in Chemistry, first in 1920 and subsequently in 1929, in both cases by German chemists. Yet, despite Bohr's deep interest in and knowledge of chemistry and of the chemical

relevance of his theory, his mind was never genuinely chemical. His approach to atomic theory was physical rather than chemical, and he tended to see chemical reasoning as merely one of several useful resources to be exploited for the higher purpose of theoretical physics. Chemical models of atoms and molecules might be useful to chemists, but if the models violated quantum physics, as they typically did, Bohr dismissed them. In some of the lectures he gave around 1920 (figure 1 shows Bohr delivering one at Yale University), he expressed the classical reductionist attitude that chemistry could be derived from atomic physics. Like Sommerfeld, he thought that a complete mathematization of chemistry was a possibility and thus the field might be a proper science according to Kant's criteria. But it would be a science subordinated to the more fundamental one of physics, not on par with it.



Figure 1. Niels Bohr talked about the correspondence principle in one of the Silliman Lectures he gave at Yale University between 6 and 13 November 1923. From the correspondence principle, he derived the selection rule that for radiative transitions in atoms, the change in the azimuthal quantum number k must be ± 1 . Bohr used that result in establishing his electron structures for the elements. (Courtesy of the Niels Bohr Archive, Copenhagen.)

In a draft document known as the Manchester memorandum, sent to Ernest Rutherford in July 1912, Bohr summarized his early ideas about atomic and molecular structure; figure 2 shows some sketches from that work. At the time he did not relate his theory to light emission or line spectra as he would do seven months later. He did discuss the mechanical stability of many-electron atoms, and he argued that in building up an electron system, it would not always be possible to confine all the electrons to a single “ring”; at some stage it would require the formation of a new, external one. In that way, Bohr established for the first time a connection between the outermost electrons of an element and its valence, a connection he found necessary in order to explain the periodic law. More than a year later, he elaborated on the suggestion in the second part of his trilogy. ¹



Figure 2. Niels Bohr's tentative structures of atoms bound into simple molecules are depicted in these drawings from the 1912 Manchester memorandum he sent to Ernest Rutherford. Note that the formulas for ozone and water are linear. In the third part of his 1913 trilogy,¹ Bohr described the molecules in words, without including drawings of them. (Courtesy of the Niels Bohr Archive Copenhagen.)

Unlike J. J. Thomson and other early architects of atomic structure, Bohr realized that the makeup of many-electron atoms could not be rigorously deduced from then-current theory. Deduction from the general principles of mechanical stability and the constancy of the angular momentum worked for the hydrogen atom, but it could not be extended to atoms with more than one electron. Bohr consequently adopted an alternative approach based on “the general view of formation of atoms . . . [and] the knowledge of the properties of the corresponding elements.” In fact, chemical inductive reasoning was more important to Bohr's determination of electron configurations than calculations based on mechanical and quantum theoretical principles.

Bohr's reliance on chemical reasoning, and his eclectic and sometimes opportunistic approach to atom building, are clearly seen in his treatment of the lithium atom. Bohr calculated the total binding energy for two configurations. One was a two-ring system that Bohr denoted (2, 1) to indicate two electrons in an inner ring and a single electron in an outer ring. The other system, (3), comprised three electrons moving on the same ring. His results were -218 eV and -240 eV, respectively, meaning that the (3) configuration is energetically favored. That conclusion is incompatible with the chemical properties of lithium. So Bohr ignored his mechanical calculations and declared the (2, 1) configuration the right one. Likewise, he had found that a mechanically stable inner ring could accommodate no more than seven electrons, a result that clearly disagreed with the known periodicity of the elements. He consequently changed the number 7 to 8—for example, assigning to sodium the configuration (8, 2, 1) rather than (7, 3, 1). As to the number of electrons in the outermost ring, he did not even pretend to base it on calculations: “The number of electrons in this ring is arbitrarily put equal to the normal valency of the corresponding element.”

The result of Bohr's eclectic method was electron configurations from $Z = 1$ to $Z = 25$ that had a

suggestive similarity to the periodic table. Realizing the tentative nature of his considerations, he cautiously avoided assigning chemical symbols to his structures or explicitly presenting them as an explanation of the periodic table. He was content to note that “it seems not unlikely that this constitution of the atoms will correspond to properties of the elements similar with those observed”—a phrase characteristic of Bohr’s language.

In the third part of the trilogy, Bohr dealt with molecules and the covalent bond, which he represented as a ring of two or more electrons common to the atoms forming a molecule. That conception of molecules had figured prominently in his Manchester memorandum, in which he presented tentative models of H_2 , O_2 , O_3 , H_2O , CH_4 , and C_2H_2 . The only molecule he discussed quantitatively and in detail was H_2 , supposed to consist of two nuclei kept together by a ring of two electrons, as shown in figure 3. A simple calculation proved that the system was mechanically stable—that is, the process $H + H \rightarrow H_2$ was exothermic, with a heat of formation of 251 kJ/mole (assuming Avogadro’s number to be 6.5×10^{23}). The calculated heat of formation was of the right order of magnitude, and Bohr’s confidence increased a few months later when US chemist Irving Langmuir informed him of improved experiments giving a result of 318 kJ/mole. With a better value of Avogadro’s constant, Bohr obtained 264 kJ/mole, only 17% below the experimental value. Alas, Langmuir’s final value of 351 kJ/mole was impossible to reconcile with Bohr’s theory.⁵ Something was wrong, but the discrepancy was not initially seen as a serious problem.



Figure 3. By letting two hydrogen atoms approach each other, Niels Bohr suggested, it was possible to understand the structure of both H_2 and helium. The drawings here show Bohr’s idea of the formation of an H_2 molecule from two H atoms, as described in works between 1913 and 1918. (Adapted from ref. 7.)

The final time Bohr discussed molecular models on the basis of his theory was in a little-known paper of 1919, in which he examined the possible existence of the H_3 molecule along the same lines he had applied to H_2 . At the time, there was some experimental evidence, both chemical

and based on experiments with positive ions, for that unusual “hyzone” form of hydrogen.⁶ From stability calculations similar to those for the H₂ molecule, Bohr concluded that H₃ probably existed and might be formed by the exothermic process $H^+ + H_2^- \rightarrow H_3$. On the other hand, he found that the H₃⁺ ion would be mechanically unstable, a result that contradicted Thomson’s discovery of H₃⁺ in 1913 and later confirmations of it. Neither Bohr nor other researchers paid much attention to the case of triatomic hydrogen, notwithstanding the anomalous experimental situation of H₃⁺ detection but no convincing evidence for H₃.

By the early 1920s, Bohr had stopped speculating about molecules, yet chemical considerations were no less important to his thinking than they had been earlier. He had abandoned his old “pancake model,” with its electrons characterized by a single quantum number and moving in planar rings around the nucleus, and replaced it with a significantly different model of atomic structure. According to the new picture, the state of an electron was characterized by two quantum numbers and designated n, k , where n is the principal quantum number and $k = 1, \dots, n$ is the azimuthal quantum number. The electrons moved in three-dimensional elliptical orbits whose eccentricity was determined by the ratio n/k . Moreover, electrons moving in outer orbits might penetrate the inner core of the atom and thereby give rise to a coupling of the revolving electrons.

In building up electron structures, Bohr was governed by what he called the Aufbau, or construction, principle: The addition of electron number p to a partially completed atom with $p - 1$ bound electrons leaves the quantum numbers of the $p - 1$ electrons unchanged, and the n quantum number of the newly added electron differs from that of the already bound electrons in the outer shell only if the atom being formed belongs to a new period of the periodic system.

Bohr presented his more sophisticated model of complex atoms in publications and meetings between 1921 and 1923, including in the Nobel lecture he gave on 11 December 1922. In Stockholm and on other occasions, he illustrated his new theory of the chemical elements by means of plates showing the electron orbits of various atoms.⁷ For example, the simple lithium atom was pictured as a helium structure made up of two crossed $1, 1$ orbits surrounded by an elliptical $2, 1$ orbit with a single electron. Figure 4 shows Bohr’s depiction of xenon. The more complex radium atom was the highlight of Bohr’s plates, with all 88 orbits meticulously drawn to scale and exhibiting a beautiful symmetry.

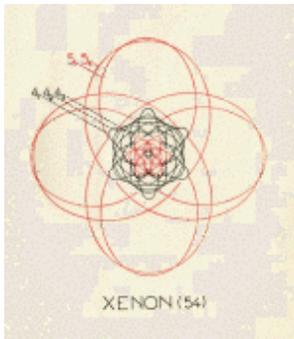


Figure 4. Detailed atomic illustrations, such as this depiction of xenon ($Z = 54$), were a feature of Niels Bohr's lectures between 1921 and 1923. According to Bohr, in the outer part (principal quantum number $n > 3$) of the atom, each of the orbits 4 1, 4 2, and 4 3 contained six electrons, and the outermost orbits 5 1 and 5 2 included four electrons each. The orbits with odd principal quantum number are red, and those with even quantum number are black. The elliptic orbits are shown closed for reasons of simplicity, but they should really be slightly open, as the ellipses slowly precess. Although drawn in two dimensions, the electrons of the real atom were supposed to move in three dimensions. (Adapted from ref. 7 .)

The plates gave the impression that electrons really moved in definite orbits, much like planets and comets move about the Sun. Although Bohr was careful to speak of the pictures as symbolic rather than concrete representations, at the time he apparently had little doubt about the reality of the electron orbits. Whereas the pictures appealed to Bohr, to the young Wolfgang Pauli they came to represent the failure of atomic models based on the illegitimate concept of electron orbits.

Confident that he had understood the basic principles of atomic architecture, Bohr suggested his own version of the periodic table arranged in horizontal groups and vertical periods, as shown in figure 5 . More importantly, for the first time he offered an explanation of the entire system from hydrogen to uranium in terms of electron configurations. Bohr's arguments implied that the unknown element with $Z = 72$ should be chemically analogous to zirconium ($Z = 40$), a prediction that was dramatically confirmed in 1923, when hafnium was discovered by means of x-ray spectroscopy in Bohr's institute. Bohr had such confidence in his theory that he undertook to write down the electron configurations of the still hypothetical transuranic elements. Thus he expected element 118 to be a noble gas with seven shells and the number of electrons in those shells given by 2, 8, 18, 32, 32, 18, and 8. Incidentally, a few nuclei of that element, provisionally named ununoctium, were manufactured in heavy-ion collisions in 2006. Modern calculations

result in an electron configuration in complete agreement with Bohr's prediction!⁸

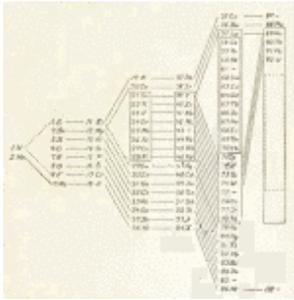


Figure 5. A periodic table as envisioned by Niels Bohr in 1923. Element 71 (Cp, cassiopeium) is today called lutetium (Lu), and element 86 (Nt, niton) is radon. Of the missing elements, number 75 (Re, rhenium) was discovered in 1925 by means of x-ray spectroscopy, the same technique that revealed the existence of hafnium in early 1923. (Adapted from ref. 7 .)

Bohr's remarkable results were not based on detailed calculations. How, then, did he obtain them? Bohr himself stressed that his theory rested on physical principles of a general nature, such as the Aufbau principle and his favorite tool, the correspondence principle. It was not, he maintained, a theory built inductively from empirical data. In reality, however, Bohr's theory of the periodic system was to a large extent based on empirical facts from chemistry and physics; those included atomic volumes, magnetic and electrochemical properties of the elements, ionization potentials, optical data, and x-ray spectral data. It is doubtful that he could have constructed his atomic configurations had it not been for his intimate knowledge of inorganic chemistry.

The empirical basis of the theory was far from clear at the time, when many physicists and chemists believed that Bohr had derived the atomic structure of the elements from quantum theory. It took some time before the theory was recognized as an artistic and somewhat opaque blend of general physical principles and empirical data, with the latter counting at least as much as the former. Dutch physicist Hendrik Kramers, who was Bohr's assistant between 1916 and 1926, wrote, "Many physicists abroad thought at the time of the appearance of Bohr's theory of the periodic system that it was extensively supported by unpublished calculations which dealt in detail with the structure of individual atoms, whereas the truth was, in fact, that Bohr had created and elaborated with a divine glance a synthesis between results of a spectroscopic nature and of a chemical nature."⁹ At any rate, Bohr's theory was short lived and soon replaced by the superior theories of Edmund Stoner and Pauli.

Bohr's attempts to extend his atomic theory to the realm of chemistry met with a mixed reception in the large community of chemists. During the first years of the theory, most chemists ignored it probably because they found it to be difficult and of little use for solving chemical problems. The theory's failure to account for the four tetrahedrally oriented valence orbitals of the carbon atom, or even for the simple hydrogen molecule, added to their suspicion that it had nothing important to offer chemistry. All the same, beginning in about 1920, many textbooks in inorganic and physical chemistry contained sections on the new atomic theory, although in most cases it was mentioned only briefly. An exception was English chemist William Lewis's 1919 textbook *A System of Physical Chemistry*, which included a detailed review of Bohr's theory based on the 1913 trilogy.

Chemists' dissatisfaction with the Bohr atom, and more generally with "the so-called quantum theory," was given voice in an address Richard Tolman delivered ¹⁰ in Toronto at the end of 1921. An eminent physical chemist and theoretical physicist (and later an authority in relativistic cosmology), Tolman had mastered the technicalities of the quantum theory of atoms, but as a chemist he found the theory unconvincing. "No chemist would be willing to think of a carbon atom as a positive nucleus with rings of electrons rotating around it in a single plane," he said, apparently unaware that this was no longer Bohr's view. On a more fundamental level, he objected to the very basis of Bohr's theory, particularly its postulates of stationary states and the mechanism of light emission in terms of quantum jumps. How could monochromatic light be emitted from an atom if there were no electrons vibrating with a corresponding frequency? Pretending to represent chemists, he stated their overall point of view as "extreme hostility to the physicists, with their absurd atom, like a pan-cake of rotating electrons, an attitude which is only slightly modified by a pious wish that somehow the vitamine 'h' ought to find its way into the vita organs of their own, entirely satisfactory, cubical atom."

Some of the objections raised by Tolman were repeated and amplified by other US physical chemists, most forcefully by Gilbert Lewis at the University of California, Berkeley, who argued that Bohr's theory was inconsistent and contradicted the fundamental principle of energy conservation. ¹¹ In spite of his criticism, he was greatly interested in Bohr's ideas. As early as 1916—at a time when Bohr was largely unknown to chemists—Lewis invited him to come to Berkeley to give a series of lectures. To Bohr's regret, he had to decline the invitation.

The main reason for chemists' dissatisfaction with the Bohr model was its inability to account for valence and the structure of molecules, in which respect they judged it inferior to the chemical models suggested by Lewis, Langmuir, and others. According to the cubical atom popular among chemists, the electrons stayed in fixed positions at the corners of a cube and the bonds between atoms were pictured as pairs of electrons common to two atoms. In that way chemists

could build up models of compounds in rough agreement with chemical facts, although only by disregarding fundamental features of atomic and quantum physics.

The central problem was that what made sense chemically was nonsense physically. It was as essential to the cubical atom that it be static as it was essential to the Bohr atom that it be dynamic. An electron moving swiftly in its orbit could not possibly occupy a fixed position in the atom, as chemists would have it. In a word, the kind of atomic models that most chemists found useful violated the standards that Bohr and his colleagues in quantum theory found necessary. While that was reason enough for physicists to dismiss static models, chemists, having their own agenda, did not feel obliged to accept the criteria for atom building that were valued so highly by their colleagues in physics. To some extent, the confrontation in the early 1920s between Bohr's dynamic atom and chemists' static one was rooted in two different cultures of science: Physicist and chemists disagreed about the criteria for what constituted a good scientific theory.

In that climate, Bohr had no patience for the models of Lewis and Langmuir, however useful they might be for elucidating chemical problems. He dismissed them as artificial and descriptive theories that lacked physical justification. The early 1920s did see several attempts to reconcile or unify static and dynamic models, typically by interpreting Bohr's model as corresponding to a static atom. In some of those hybrid models, the covalent bond was pictured as one or two electrons orbiting elliptically around two nuclei rather than circulating between them. Bohr was aware of the popularity of what one chemist called the "Rutherford-Bohr-Lewis-Langmuir atom," but he denied that a reconciliation was possible within the framework of the quantum theory of the time.

Whereas most chemists agreed that Bohr's model was useless in the areas of valence and molecular structure, they were more positively inclined to his theory of the periodic system. According to Walther Nernst, a pioneer of physical chemistry and a 1920 Nobel laureate, the discovery of hafnium proved that Bohr's theory was essentially correct. Furthermore, Bohr's quantum theory proved valuable for the study of molecular spectra in the branch of chemistry that came to be known as chemical physics. That interdisciplinary field evolved into an important branch of research in the early 1920s, to a large extent guided by the ideas of atomic and molecular structure developed by Bohr and other quantum physicists.

Although Bohr's original and most ambitious project of establishing a common theory for atoms and molecules—one that would be of equal significance to physics and chemistry—turned out to be a failure, it was not without fruitful consequences. In a sense, his ambitions would be fulfilled with the emergence of quantum mechanics and its extension into quantum chemistry in the late 1920s.

On the other hand, the tensions between physicists and chemists were only aggravated with the advent of quantum mechanics. Not only was the new theory incomprehensible to most chemists because of its abstract nature and mathematical complexity, but it also seemed to share the impotence of the old Bohr theory with regard to chemical applications. The first breakthrough in quantum chemistry—the fully quantum mechanical description of the H₂ bond presented by Walter Heitler and Fritz London¹² in 1927—was the product of physicists working in the culture of avant-garde theoretical physics, with almost no knowledge of or concern for problems of structural chemistry. London had no respect for chemists' models of valence. As he arrogantly said in a letter to Heitler, "The chemist is made out of hard wood and he needs to have rules even if they are incomprehensible."¹³

Only with Linus Pauling's important work in the 1930s did it become clear that the success of quantum chemistry would depend crucially on chemical facts and modes of reasoning. The new generation of quantum chemists had to liberate themselves from physicists' modes of thought and create a new interdisciplinary framework for their science. Many of the founders of quantum chemistry had visited Bohr's institute in the 1920s, but when quantum chemistry took off, neither Bohr nor his collaborators followed up on the development.

Yet Bohr was well acquainted with the new discipline and aware of its significance. When the young Erich Hückel went to Copenhagen in 1929 to do research in quantum mechanics, it was Bohr who directed him to the Heitler–London theory and suggested that he look at the double bond.

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