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Walther Nernst and the Development of Physical Chemistry

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Nernst, Walter · physical chemistry · thermodynamics · science history

Walther Nernst (Figure 1), one of the pioneers of physical chemistry, was born on the 25th June 1864 in Briesen (formerly West Prussia). To commemorate his 100th birthday, a plaque was put up at his former premises at the Humboldt University, Berlin (Figure 2) to honor his most significant scientific achievement, the discovery of the 3rd law of thermodynamics. The life and work of this outstanding scholar has already often been described extensively,[1–4] so the present Essay concentrates essentially on his work in his time and that subsequent to the development of physical chemistry.

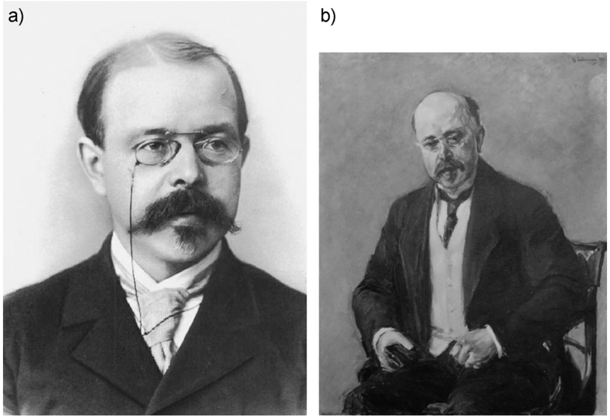

Figure 1. Walther Nernst (1864–1941): a) in his early years and b) in a picture by Max Liebermann (painted 1912, lost 1945).

IM JAHRE 1905 ENTDECKTE
WALTHER NERNST
IM VERLAUF SEINER IN DIESEM
SAALE GEHALTENEN VORLESUNG
DEN 3. HAUPTSATZ
DER THERMODYNAMIK.
DIE HUMBOLDT-UNIVERSITÄT
GAB 1964, IM JAHRE
DES 100. GEBURTSTAGES
DES GROSSEN GELEHRTEN,
DIESEM HÖRSAAL DEN NAMEN
WALTHER-NERNST-
HÖRSAAL

Figure 2. Commemorative plaque at the former institute of Walther Nernst at the Humboldt University Berlin.

How was the discipline when Nernst prepared himself to think about his future prospects? The famous physiologist and cofounder of the physical society of Berlin (the predecessor organization of the German Physical Society) Emil du Bois-Reymond should be quoted here. He explained in a speech to the Prussian Academy of Science on 29th June 1882:[5] “*In contrast to modern (i.e. actual) chemistry, one can name physical chemistry as the chemistry of the future … It applies indeed to modern chemistry at its peak, what Kant said of chemistry in his time. It is a science, but not science; not in this sense, in which there is actually science, particularly in the sense of the understanding of nature advanced by mathematical mechanics … Science in its highest human sense would first be chemistry if we understood the tensions, velocities, unstable and stable equilibrium states of particles causal from nature as the movements of the celestial bodies. The astronomy is certainly more advanced than chemistry, which in observant aceticism remains at a stage, still under the astronomy in Kopernikus’ and Keplers times*”.

At that time, there were already attempts to use the methods of physics in chemistry problems. Only the chemist Bunsen and the physicist Kirchhoff in Heidelberg are named here. The leader of the newly emerging discipline of physical chemistry was Wilhelm Ostwald (Figure 3), who was 11 years older than Nernst and established a large school first in Riga and then from 1887 in Leipzig. Apart from numerous fundamental discoveries, which even until today are partly associated with his name, he was able to explain catalysis as a phenomenon of reaction velocity (i.e. kinetics) and was thus awarded the Nobel Prize for Chemistry in 1909. Besides him, the Dutchman of about the same age, Jacobus Henricus

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Figure 3. Wilhelm Ostwald (1853–1932).

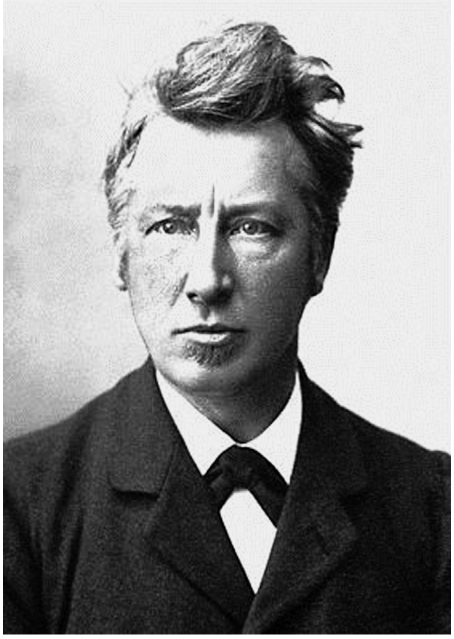


Figure 4. Jacobus Henricus van't Hoff (1852–1911).

$$\mathrm{A} + \mathrm{B} \rightleftharpoons \mathrm{C}$$

$$\text{Equilibrium:} \quad \frac{[\mathrm{C}]}{[\mathrm{A}]\cdot[\mathrm{B}]} = K(p,T)$$

$$\frac{\partial \ln K}{\partial T} = \frac{\Delta H}{RT^2} \quad \text{Van 't Hoff}$$

Figure 5. The van't Hoff Equation for the temperature dependency of the equilibrium constant *K* of the enthalpy of reaction Δ*H*.

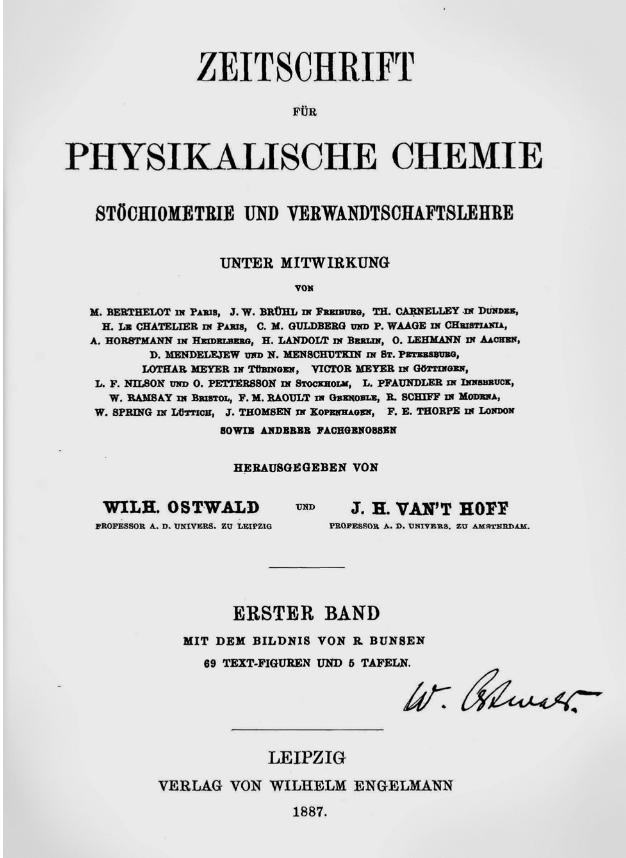
van't Hoff (Figure 4) must primarily be named. He obtained the first chemistry Nobel Prize in 1901 for his work on chemical dynamics and osmotic pressure, and was professor in Berlin from 1886. Furthermore, his name is known by every chemistry student from the equation named after him on the temperature dependence of the constant of a chemical equilibrium (Figure 5). Here we encounter for the first time an attempt to quantification of a "stable (chemical) equilibrium" stipulated by du Bois-Reymond: The enthalpy of the reaction Δ*H*, obtainable by calorimetric measurement, enables determination of the temperature dependence of the equilibrium constant *K*. Integrating the equation leaves an unknown integration constant, so that an absolute determination of *K* by this way is not possible (i.e. solely from calorimetric data). The discovery of the 3rd law by W. Nernst was first necessary for this.

The triumvirate of the founding fathers is completed by the Swede Svente Arrhenius (Figure 6), who was awarded the Nobel Prize in 1903 for his work on the dissociation of salts into ions in solution. At the time of his dissertation (1884) this idea was revolutionary, and he didn't at first find any proper appointment, but was able to get to know the important laboratories in Europe with a travel scholarship. Thus, he also met Walther Nernst, who was working with Friedrich Kohlrausch in Würzburg in 1887 after study visits in Zürich, Berlin, and Graz (Figure 7). In the summer of 1887, the two friends travelled to Graz again to the institute of Ludwig Boltzmann (Figure 8), where Nernst had, together with his assistant Albert von Ettinghausen, discovered the effect named after the two of them, which is based on the fact that (similar to the Hall effect) a metallic conductor with a heat

Figure 6. Svante Arrhenius (1859–1927).

Figure 7. The group of F. Kohlrausch in Würzburg in the spring of 1887. Standing, from left: Heydweiler, Rasch, Arrhenius, Nernst. Sitting, from left: Palazzo, F. Kohlrausch, Sheldon.

Figure 8. The group of Ludwig Boltzmann at the University of Graz in autumn 1887. Standing, from left: Nernst, Streintz, Arrhenius, Riecke. Sitting, from left: Aulinger, von Ettingshausen, Boltzmann, Klemencic, Hausmanninger.



ZEITSCHRIFT
FÜR
PHYSIKALISCHE CHEMIE
STÖCHIOMETRIE UND VERWANDTSCHAFTSLEHRE

UNTER MITWIRKUNG
VON
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LOTHAR MEYER IN TÜBINGEN, VICTOR MEYER IN GÖTTINGEN,
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HERAUSGEGEBEN VON
WILH. OSTWALD UND J. H. VAN'T HOFF
PROFESSOR A. D. UNIVERS. ZU LEIPZIG PROFESSOR A. D. UNIVERS. ZU AMSTERDAM.

ERSTER BAND
MIT DEM BILDNIS VON R. BUNSEN
69 TEXT-FIGUREN UND 5 TAFELN.

W. Ostwald

LEIPZIG
VERLAG VON WILHELM ENGELMANN
1887.

Figure 9. The title page of the first issue of the *Zeitschrift für Physikalische Chemie* published by van't Hoff and Ostwald in January 1887.

flux passing through it builds up a voltage under the influence of a magnetic field. He obtained his doctorate with this work in Würzburg in the same year. Before that, he had also got to know Wilhelm Ostwald in Graz, about whom Arrhenius had already raved. Ostwald had just obtained a call to the only chair for Physical Chemistry existing at that time in Leipzig and offered Nernst a position as assistant there, which he gladly accepted. Nernst was responsible for the practical training, but as there were initially only two participants, he had plenty of time for his own research.

At the same time, Ostwald and van't Hoff founded together the *Zeitschrift für Physikalische Chemie* (Figure 9), the oldest publication medium of this discipline, which continues to the present day.

Triggered by the works of Arrhenius and van't Hoff, the interests of Ostwald then focussed mainly on the investigation of ions and their electrochemical properties, and as such he also wanted to guide his assistant Nernst in this field. In his memoirs he noted:[7] "*naturally I am not entitled to claim that the first seed for that chain of thought resulted from this conversation in the corner room of the old institute. Indeed, I had then the impression for Nernst it concerned ideas which he did not want to follow without an inner resistance.*"

Ostwald suggested initially to Nernst to determine the heat of formation of mercury salts to verify a relationship between heat of formation and electromotive force (EMF) in a galvanic cell previously postulated by Helmholtz.[8] The existence of the ions still remained the dominant topic, and the influence of Ostwald on Nernst is even significantly further emphasized by the following publication ("*Über freie Ionen*" ("About Free Ions"),[9] which was published under both names. However, Nernst (who intrinsically felt himself to be a physicist) had meanwhile found such an interest in electrochemistry that he created the thermodynamic fundamentals of this subject in a monumental work[10] and thus qualified as a professor ("habilitation") at the age of 25. The Nernst Equation about the concentration dependence of EMF must surely be known, even today, by every chemistry student.

Nernst now emerged out of the shadows of Ostwald and went to Göttingen, where in 1890 he was, alongside Paul Drude, assistant at the Physical Institute and one year later associate professor for physical chemistry.

Meanwhile, Nernst had achieved a certain illustriousness, so that others were also interested in him at other places. To avoid his exodus from Göttingen, he was appointed full professor there in 1894, with the promise that an Institute for Physical Chemistry and Electrochemistry (the first of its kind!) would be established for him. In the same year, the German Electrochemical Society was founded by Ostwald, van't Hoff, Nernst, and others, which was renamed the German Bunsen Society for Applied Physical Chemistry in 1902 and since 1936 is today's name without the term "Applied". Nernst was its first President from 1905 to 1908, was later honorary member, and was awarded with the Bunsen Commemorative Medal. A bit later (1898), Ostwald also obtained his own Institute for Physical Chemistry in Leipzig—until then he was the director of the 2nd Chemistry Institute. Following that, chairs for Physical Chemistry and Electrochemistry were established at numerous universities; in 1911 followed the founding of the Kaiser Wilhelm Institute for Physical Chemistry and Electrochemistry in Berlin-Dahlem with Fritz Haber as the first director, which since 1953 carries his name as the Institute of the Max Planck Society. In 1919 the University of Munich came to an end, as the ruler there, Adolf von Baeyer, did not want any other king besides him. Its own institute building was then inaugurated in 1932.

In his official speech[11] at the opening of his institute at Göttingen, Nernst said: "*The current day constitutes an important demonstration in that between two rather hitherto separated sciences a close reunification has occurred.*" He estimated the period of the separation to around 1835–1885: The separation occurred as the methods and concepts of both subjects distanced themselves from each other, while for him the reunification started in 1885 when van't Hoff's famous work about dissolved substances and Ostwald's first volume of his great textbook were published. However, he was not successful in a clear definition of the two disciplines, mainly because the existence of atoms was still only a hypothesis at this time.

The research work during the years at Göttingen extended over different areas of physical chemistry, but was principally concentrated on electrochemistry. In this regard, experiments on the conductivity of solids (oxides) were carried out, which led to the invention of the Nernst lamp. A mixture of various oxides is conductive at higher temperatures and eventually glows and emits bright light. The lamps based on this principle were produced in large quantities (Figure 10) and served, for example, for the lighting of the Paris world exhibition in 1900. One disadvantage was the necessity of preheating, so that the end of this technology neared when it became possible to produce high-melting metal wires. The final fatal blow came from a former doctoral student of Nernst, the American Irving Langmuir (Nobel Prize 1932), who filled the light bulbs with a protective gas. Nernst had, however, such business acumen to previously sell his patents as a whole to AEG for a horrendous sum—one spoke of a figure of 1 million gold marks! He became a wealthy man by this, which was assisted by marriage to a rich heiress, and he was able to afford, for example, the first automobile in Göttingen and also various country estates.

He then used the automobile to move his family to Berlin, where he followed a call for the professorship for Physical Chemistry as the successor to Hans Landolt.

Figure 10. Fabrication of Nernst lamps around 1900 at AEG. Source: Ref. [4], p. 97.

... The Stable Equilibrium Positions

The calculation of the constant K for a chemical equilibrium on the basis of calorimetric data (which are obtained by temperature measurements) represented not only a challenge for a quantitative theoretical system but was also of enormous importance for the emerging area of technical chemistry. The temperature dependency of the equilibrium constant K on the reaction enthalpy ΔH from the van't Hoff Equation according to Figure 5 had already been discussed, in which the quantitative information with an unknown integration constant was limited. This can be explicitly represented by the relationship $RT\ln K = -\Delta G^\circ$, where ΔG°, the free enthalpy of a reaction (under standard conditions), consists of the enthalpy of reaction ΔH° and the entropy of reaction ΔS°. The entropy $S(T)$ is calculated from the molar heat capacity C_p from $S = \int_0^T \frac{C_P}{T} \mathrm{d}T + S_{T=0}$. Nernst ascertained now that the difference ΔS of the entropy between the product and reactants disappears upon approaching absolute zero, that is $\lim_{\Delta T \to 0} \Delta S = 0$. Nernst came to this conclusion because he found that the experimentally determined differences in the molar heat capacities for $T \to 0$ approach zero. That is the basis of Nernst's heat theorem, which now enabled calculation of the chemical equilibria on the basis of calorimetric data (heat of reaction and molar heat capacity). This heat theorem was extended by Max Planck to the 3rd law, whereupon the entropy of ideal crystalline solids takes a value of zero at absolute zero. Considering the statistical theory of the entropy $S = k\ln W$ by Ludwig Boltzmann, this deduction does not seem surprising, but at the time of the construction of Nernst's heat theorem, these ideas were not at all generally accepted; among others, Oswald also belonged to its opponents.

To check the validity of his heat theorem, Nernst was naturally interested in experimental data about chemical equilibria in the gas phase. In doing so, he came across a reaction which attracted considerable attention at that time: the formation of ammonia from the elements: $N_2 + 3H_2 \rightleftharpoons 2NH_3$. At the beginning of the 20th century, it was becoming clear that the natural deposits of nitrogen fertilizer (Chile saltpetre) would soon be exhausted if it wasn't possible

Figure 11. Fritz Haber (1868–1934).

to transform the inert N_2 molecule by “nitrogen fixation” into a reactive compound (“bread from air”). In Karlsruhe, the associate professor there Fritz Haber (Figure 11), motivated by an enquiry from industry, had undertaken a series of experiments in which barium and calcium were converted interchangeably into nitrides and hydrides, during which ammonia is formed.[13] The yields obtained were so unsatisfactory that Haber did not think of a technical application, which he illustrated in detail in his book published at that time.[14] However, the data for the equilibrium given by him did not correlate with the calculations of Nernst, who expected even lower ammonia yields and who now performed his own experiments at higher pressures and (thus higher) yields. Following the presentation of these results at the Bunsentagung in 1907,[15] a heated debate took place between the Geheimrat of Berlin (who at that time was also the first president of the Bunsen Society) and the virtually unknown associate professor from Karlsruhe, in which Nernst said: “*It is regrettable that the equilibrium is shifted to the side of the much lower yields, as one up to now has assumed, according to the highly incorrect figures of Haber, for one would really have thought to synthetically produce ammonia from nitrogen and hydrogen. But now the conditions are much more unfavorable.*”

The matter was thus settled for Nernst, but not for Haber. Together with a talented young co-worker, Le Rossignol, he set about to build a new apparatus which also functioned at higher pressures. He was thus able to come near to the values of Nernst for the equilibrium.[16] On the 2nd July 1909 they finally succeeded to produce ammonia in this way through further improvements and choice of a suitable catalyst,[17] from which Carl Bosch and Alwin Mittasch developed the industrial Haber–Bosch process, which went into operation in September 1913 and quickly revolutionized the production of fertilizer and explosives. Fritz Haber, who in actual fact owed this success to the controversy with Walther Nernst, was awarded the Nobel Prize in 1918 for this, while Nernst in 1916 in a letter to BASF wrote: “*unfortunately I then held the view that it is technically difficult to make*”. Nernst had to wait a while for the Nobel Prize, until he obtained it in 1920 (awarded 1921) in “recognition for his thermochemical work”. The delay was probably due to the fact that the relationship between the earlier friends Arrhenius and Nernst was troubled since about 1900.

As mentioned above, the pursuit of the heat theorem of Nernst led to the problem of the molar heat capacity at low temperatures. According to classical theory, this should be independent of temperature, while the experiments suggested a decline to zero. A theoretical explanation for this was first suggested by Albert Einstein in 1907 based on the quantization of oscillations in solids.[18] He assumed here that all atoms oscillate with the same frequency and transitions are always associated with the energy $h\nu$. This localized view was later extended by Debye[19] by using a model with coupled oscillations, whereby considerably better agreement with the experimental values at very low temperatures were able to be achieved.

Thermochemistry at low temperatures also formed one of the central issues of the following work at the Nernst institute. The satisfactory description of the specific heat of solids at low temperatures by taking as a basis the quantum hypothesis introduced by Planck (initially considered by him to be only an auxiliary construction) prompted Nernst to propose a conference with the leading scientists occupied with this question. This took place in Brussels in October 1911 by invitation of the Belgian industrialist Solvay (Figure 12). In his substantial contribution,[20] Einstein firstly placed the result of his theory in the context of Planck's radiation formula and also pointed out the deviations to the experimental data, which he traced back to the assumption of a single oscillation frequency. He also noted that Nernst “*extricated all appertaining results here from their theoretical shadows*”, although both did not consistently have the same opinion. However, he also added: “*The difficulties which are opposed to a satisfactory theory of these fundamental processes seem insurmountable at the moment*”. For him, the only assured foundations of a new theory existed in the conservation of energy and in Boltzmann's definition of the entropy by probability.

... The Tensions

Now the atomic structure of matter had to be explained, which was still very contentious at the beginning of the 20th century. Only the discovery of the interference of X-rays in 1912 by Max von Laue (Nobel Prize 1914) established the possibility of determining the structure of substances up to complicated biological molecules. The arsenal of experimental methods has meanwhile enormously broadened by the development of further techniques, and nowadays this question can principally be considered solved.

The explanation of the chemical bond and the energy state in molecules and solids had to wait for the development of

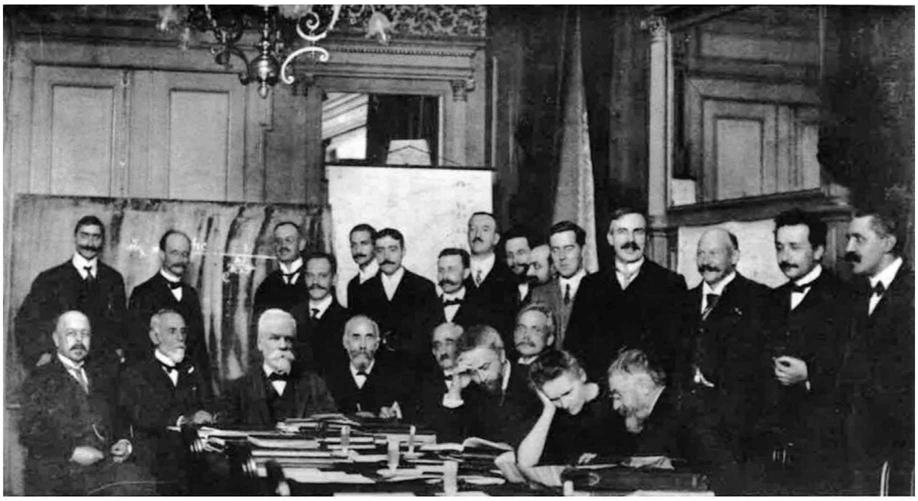

Figure 12. Group photo of the 1st Solvay congress 1911 in Brussels. Standing, from left: Goldschmidt, Planck, Rubens, Sommerfeld, Lindemann, de Broglie, Knudsen, Hasenöhrl, Hostelet, Herzen, Jeans, Rutherford, Kammerlingh Onnes, Einstein, Langevin. Sitting, from left: Nernst, Brillouin, Solvay, Lorentz, Werburg, Perrin, Wien, Mme. Curie, Poincaré.

quantum mechanics as asked for by Einstein. The theoretical treatment succeeded principally by solving the equation of wave mechanics developed by Erwin Schrödinger (Nobel Prize 1933). In the subdiscipline developed as quantum chemistry, there were numerous approaches designed for approximately solving the Schrödinger equation. The density functional theory (DFT) developed by Walter Kohn (Nobel Prize 1998) proved to be particularly efficient. One year before Schrödinger, Werner Heisenberg was awarded the Nobel Prize for developing an alternative (but equivalent) quantum theory, "*whose application led amongst other things to the discovery of the allotropic forms of the hydrogen molecule*". This is based on an early experimental confirmation of the new quantum mechanics: According to this, both protons in the H_2 molecule each possess a nuclear spin which can be aligned either parallel (ortho-H_2) or antiparallel (para-H_2) to each other. The coupling of this angular momentum with the molecular rotation causes both allotropic forms to occur either with only even- or odd-numbered rotational quantum numbers. The temperature-dependent different occupations of these states are reflected in corresponding small differences in the molar heat capacity and thermal conductivity. The young scientists Karl-Friedrich Bonhoeffer and Paul Harteck succeeded in experimentally verifying this at the KWI for Physical Chemistry in Berlin-Dahlem.[21]

Meanwhile, a large arsenal of methods has been developed to experimentally detect the energy states in molecular systems. Their detailed explanation would, however, go beyond the scope of this Essay.

... The Velocities

The investigation of the velocities (rates) of chemical reactions was mainly advanced by the successor of Nernst, Max Bodenstein (Figure 13). On the experimental side, the development of methods to detect ever shorter half-times followed, as illustrated in Figure 14: while the limit in Nernst's times was at about 1 s, the development of relaxation methods by Manfred Eigen (Nobel Prize 1967) enabled the determination of rate constants of the fastest (i.e. diffusion limited)

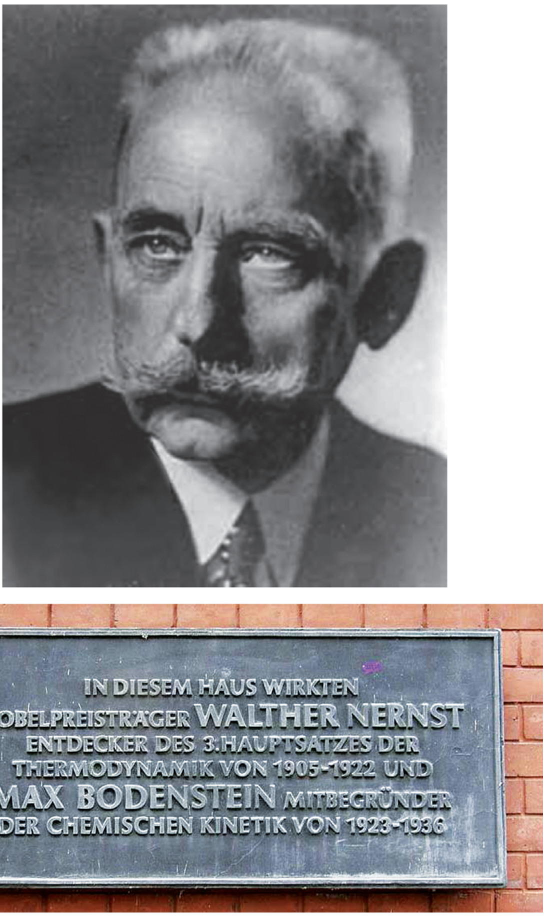

Figure 13. a) Max Bodenstein (1871–1942) and b) commemorative plaque for Nernst and Bodenstein at the former Berlin Institute, Bunsenstrasse 1.

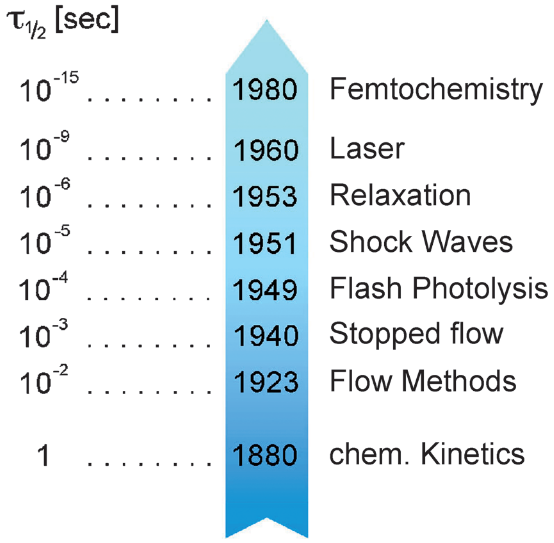

Figure 14. Chronology for the development of experimental methods to determine the half-lives $\tau_{1/2}$ of chemical reactions.

reactions in solution, and in the 1980s Ahmed Zewail (Nobel Prize 1999) succeeded by using laser techniques to reach down into the area of femtoseconds (10^{-15} s), through which the movements of atoms could be directly detected. Nowadays, the movements of electrons can even be followed with attosecond techniques (10^{-18} s). On the theoretical side, the calculation of the theoretical energy of the simplest reaction, the linear conversion $H + H_2 \rightarrow H_2 + H$ as a function of both relevant distances (Figure 15) followed after the development of quantum mechanics, and consequently the formulation of the theory of the transition state by Henry Eyring and Michael Polanyi,[22] also at the KWI for Physical Chemistry, Berlin. On the basis of this theory, the fruitful area of reaction dynamics was developed, in which enormous progress could be made in understanding elementary chemical reactions both experimentally (e.g. by molecular beam and laser techniques) and also theoretically.

… The Unstable Equilibria

The rules for stable thermodynamic equilibria in closed systems are given by the laws of thermodynamics, with which Nernst with his heat theorem had supplied the corner stone. In addition, open systems in stationary conditions (which means constant external conditions and inflow and outflow rates) as "flow equilibria" can, however, show definite space-time pattern formation and/or temporal oscillatory behavior. Observations of this sort had already been reported since the 19th century. The Belousov–Zhabotinsky reaction gained a certain fame here.[24] As an example, the periodic kinetics in the anodic dissolution of copper in hydrochloric acid investigated by Bonhoeffer with his doctoral student Heinz Gerischer will be pointed out.[25] This study is quoted here

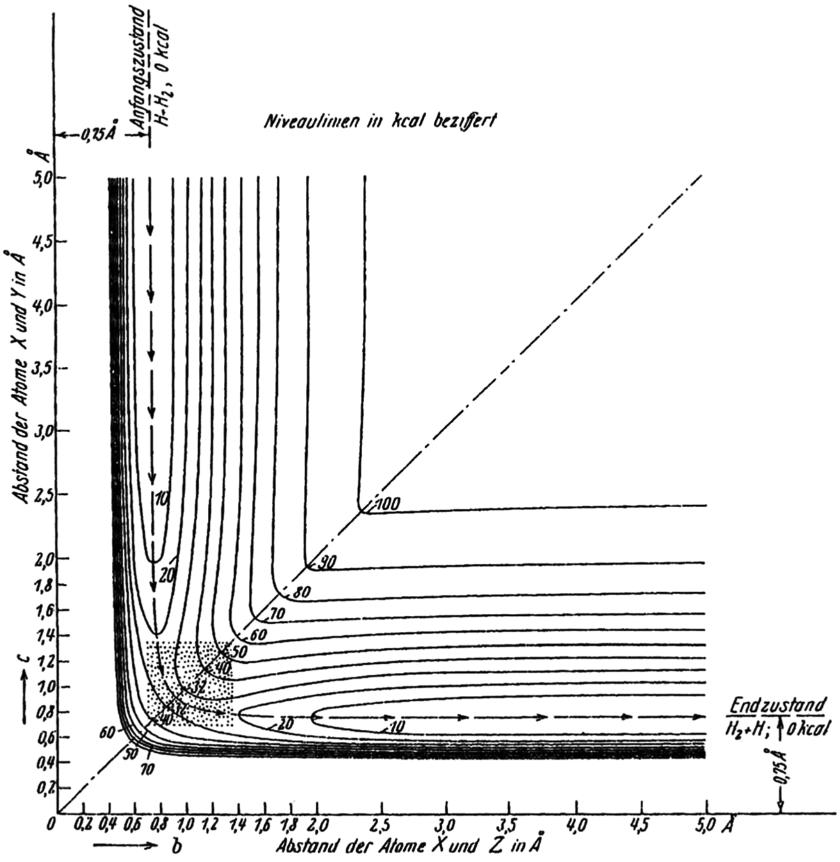

Figure 15. Hypersurface potential for the reaction $H + H_2 \rightarrow H_2 + H$ as a function of both distances b and c according to Eyring and Polanyi.[22]

because Bonhoeffer was a doctoral student of Nernst and the author of this Essay was a doctoral student of Gerischer, which illustrates the line of descent. Such phenomena are also particularly characteristic for living nature, and in a much regarded book "What is Life?", Erwin Schrödinger in 1944 posed the question whether the known physical laws are also sufficient to explain these phenomena. He was unable to deliver a satisfactory answer to this and yet established a milestone for the development of molecular biology with this book. A first theoretical model for pattern formation in nature (morphogenesis) was suggested by the famous English mathematician Alan Turing, whereafter chemical reactions interact with transport processes (diffusion) without assuming that new types of laws would be necessary.[26] The complete theoretical basis for such structure formation in open systems far from the thermodynamic equilibrium was then developed by the Belgian Ilya Prigogine (Nobel Prize 1977) with the term dissipative structures.[27] The mathematical description comprises the coupling of nonlinear partial differential equations for the kinetics with the diffusion of species taking part in the reaction, whose solutions reproduce the observed spatiotemporal self-organizations.[28]

Epilogue

After being awarded the Nobel Prize, Nernst retreated more and more from physical chemistry. He was rector of the University of Berlin, president of the Physikalisch-Technischen Reichsanstalt for a short time, and then returned to a physics professorship, as his previous chair was now taken by Bodenstein. He occupied himself with astrophysics and the development of an electric piano. A photo from the later years (Figure 16) shows him as "preceptor" in the circle of other Nobel Prize winners. In 1932 he was given emeritus status and retired to his manor Zibelle (Oberlausitz), where he died on the 18th of November 1941.

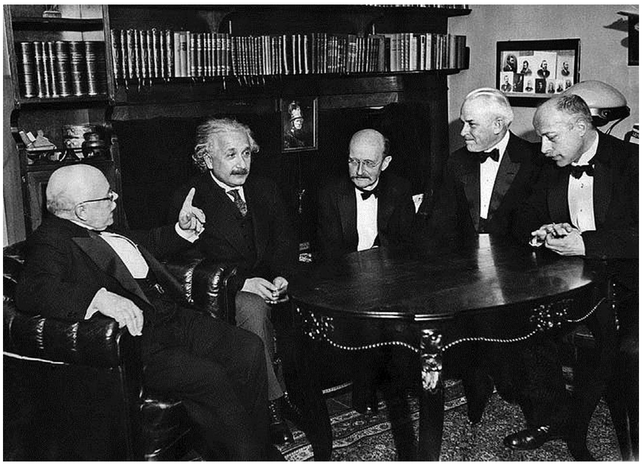

Figure 16. Walther Nernst in 1931 among other Nobel Prize winners. From left: Nernst, Einstein, Planck, Millikan, and von Laue.

The list of challenges for physical chemistry established by du Bois-Reymond at the start of his scientific career can—not least by the vital contributions of Nernst—be principally regarded as fulfilled, so that chemistry as a science can be valid "*in the sense of the understanding of nature advanced by mathematical mechanics*", where numerous scientists have worked on its complicated and complex structure and will continue to do so. A quote from the great Hermann von Helmholtz from the same time cautions, however, for modesty: "*The aim of the research of nature is to resolve the initially strange nature confronting us, by the logical force of law… However the field, which can be submitted to the unconditional ruling of perfect science, is unfortunately still very narrow.*"

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