Pierre Maurice Marie Duhem: A Polemical Scientist

Jaime Wisniak

Department of Chemical Engineering, Ben-Gurion University of the Negev, Beer-Sheva, Israel 84105,
wisniak@bgumail.bgu.ac.il

Abstract: Pierre Duhem was a multifaceted and prolific scientist active in thermodynamics, physics, history, and philosophy. His rigid and noncompromising attitude, together with deep religious feelings at a time when France was riding a very strong wave of anticlericalism, led to his being unable to teach in Paris and to academic exile in the provinces. He was a prolific writer and he left his name on many equations such as the Gibbs–Duhem and Duhem–Margules equations. We present here a general picture of his life and activities against the political climate in France as well as a discussion of some of his most important contributions to thermodynamics.

Life and Career

Pierre Maurice Marie Duhem was born in Paris on Sunday, June 9, 1861, the son of Pierre-Joseph Duhem, of Belgian origin, and Marie-Alexandrine Fabre, whose family originated from the town Cabesprine, near Carcasonne. At the age of seven he began his studies in a private school and in 1872, at the age of ten, he continued and completed his education at a Catholic boarding school, the Collège Stanislas in Paris. His father, worried by the financial burden of a private school and aware of the better prospects of private-school graduates in admissions to the best universities (e.g., the Sorbonne, Polytechnique, and École Normale) had wanted to send him to a state school. Nevertheless, his mother's opinion had prevailed, and Collège Stanislas was chosen. Duhem's mother was particularly sensitive to the negative anticlerical and secularist influences that might sway her son from his orthodox religious upbringing [1–3]. As described below, at that particular time free thinking was increasingly felt in state schools and public forums, while legal support of ecclesiastical influence had begun to erode.

The years at Stanislas were difficult years in Paris, with the Franco–Prussian War raging until the armistice in February 1871. The year 1872 brought personal tragedies to the Duhem family: a diphtheria epidemic killed Duhem's young sister Antoinette-Victorine and his recently born brother Jean, leaving only Pierre and Antoinette's twin sister Marie-Julie (who eventually became a nun). At the age of 16 (1877) Duhem contracted a severe rheumatism that plagued him for the rest of his life. Owing to his sickness he could not participate in the 1881 competitive entrance exams to the École Normale; his participation was postponed for one year. He took additional courses at Stanislas, and in July 1878 he became *bachelier-ès-lettres* and in July 1879 *bachelier-ès-sciences* [1]. As a result of his temporary inability to enter the École Normale, Stanislas hired him as an assistant teacher. According to his sister [3], Duhem was the best student in his class during the five-year studies, although the *Annuaires* of the Collège give a different picture, ranking him fourth in a class of 34. In 1882 Duhem passed the entrance examinations to the École Normale Supérieure, in first place among 471 applicants [1–3]. In 1885 he graduated but remained at the École for one more year studying additional subjects and serving as an assistant in physics.

Before continuing, it is important to recall the events that took place during that period in the history of France [4] because eventually they would play a crucial role in the casting of Duhem's personality. In September 1870 the Germans conquered Sedan and took the French Emperor Napoléon III and thousands of soldiers prisoner. Duhem's mother fled Paris, taking young Pierre and his two sisters to an uncle living in the town of Chateudun. The family fled again to Bordeaux after Germans besieged and captured the town [1, 2]. On September 4, 1870, after the defeat in Sedan, a popular uprising toppled the government of the Second Empire and the provisional government of national defense eventually signed the armistice in February 1871. One stipulation of the armistice called for the prompt election of a national assembly with authority to negotiate a definitive treaty of peace. That election, held on February 8, produced an assembly strongly dominated by monarchists. The National Assembly convened first in Bordeaux and eventually moved to Versailles. No sooner had it arrived there when it was confronted by a major civil war—the rebellion of the Paris Commune. According to Karl Marx, the Commune should be considered the first great uprising of the proletariat against its bourgeois oppressors. Although there was a class-struggle element in the episode, its main reason was the Parisian outrage by the action of rural France in electing a monarchist assembly committed to what they regarded as a dishonorable peace and transferring the capital to Versailles rather than to Paris. On March 26, Parisians elected a council that adopted the traditional label "Commune of Paris." Its membership ranged from radical republicans of the Jacobin and Blanquist variety to socialists of several different sorts, who favored a decentralized federation of self-governing communes throughout France. The Communards were eventually defeated by the forces of the National Assembly in a series of bloody battles, followed by reprisals, and acts of vengeance. Tens of thousands of Communards were killed or executed and another large number deported to the penal islands. Around 1880 the republicans regained control of the Legislature of the Third Republic and stood for a strong centralized regime, intransigent anticlericalism, an assertive nationalism in foreign policy, a revision of the constitution to prune out its monarchical aspects, social reforms as labor laws,

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Figure 1. Photographs of Duhem. Top: Duhem (second from the left) during a doctoral examination. The Clausius–Clapeyron equation appears on the blackboard.

and a graduated income tax. Eventually, primary education became free, compulsory, and secular, with religious teaching in the public schools replaced by civic education; a strong anticlerical bias thenceforth marked French public education. A new extreme moral and political low was reached in 1894 with the Dreyfus affair. The new-left majority that arose afterward retaliated by bringing the army under more rigorous civilian control and by embarking on a new wave of anticlerical legislation. Most religious orders were dissolved and exiled, and in 1905 a new law separated church and state, thus liquidating the Concordat of 1801 [4]. During this event Duhem, consistent with his strong religious beliefs, sided with the Army and against Dreyfus.

At the École Normale Duhem received a license in mathematics and another in physics at the end of the 1883–1884 academic year. In Duhem's final year Louis Pasteur asked Duhem to join his laboratory as head of bacteriological chemistry of the Institut Pasteur, but Duhem refused because he wanted to be a physicist. His student paper published in the school's periodical used the Gibbs function (which Duhem called "thermodynamic potential") to analyze the behavior of a voltaic pile and signaled the initiation of a life-long animosity with Marcellin Berthelot.[1] In his paper Duhem claimed that his conclusions should be considered the third principle of thermochemistry. To understand the root of the problem it must be remembered that at that time Berthelot was the undisputed star of French science in general and of thermochemistry in particular. Furthermore, the expression "third principle of thermochemistry" was associated with certain conclusions (later proved not to be general) of Berthelot. One could argue that at that time Duhem was too young to realize the troubles that he was buying with his bold and head-on criticism of a scientific and political figure, but this attitude would characterize him throughout his whole life. The first blow occurred during the 1884–1885 academic year when he presented a thesis in physics for his doctorate. His thesis, *Le Potentiel Thermodynamique et ses Applications à la Mécanique Chimique et à l'Étude des Phénomènes Électriques*, was rejected by a panel of three scholars. The speculation was that the panel, chaired by Gabriel Lippmann (1845–1921, professor of physics at the Sorbonne, 1908 Nobel Prize for his work on color photography), made a political decision [5]. The evaluation report by Lippmann was wholly negative, arguing that the author (Duhem) had not only misunderstood the true meaning of Clausius' formula of entropy but also ignored substantial reservations made to its validity by Clausius himself. In addition, Lippmann claimed that all the conclusions drawn by Duhem were too vague to be submitted to verification and that they were applicable to an infinite number of different formulas [1]. What makes Lippman's negative opinion questionable is that the thesis contained the formula that eventually became known as the Gibbs–Duhem equation [1, 5]. A version of the thesis was published as an almost 250-page book by the end of 1886 by the prestigious French scientific publisher A. Hermann [6]. Two years later Duhem defended another thesis in applied mathematics, *Sur l'Àimantation par Influence*, and received his doctorate in mathematics from the Sorbonne in October 1888. The three-member jury panel that judged the thesis included Henri Poincaré. Interestingly enough, the second thesis was based on the same concept of thermodynamic potential defined in the one rejected, though applied to electromagnetics with which, according to Lippmann, it had nothing to do [1]. The 140-page thesis was published by the famous publishing house Gauthier-Villars (the same publishers that in 1824 had published Carnot's epoch-making brochure elucidating the principles of conversion of heat into work).

Berthelot had said, "This young man will never teach in Paris." Berthelot's influence was so strong that the Ministry of Public Instruction respected his recommendation even after Berthelot's death in 1907. Berthelot's edict came true, and Duhem spent his entire life and academic career in provincial universities far from Paris, the center of academic life in France [5]. Duhem's conservative and religious views were certainly significant factors in this harsh situation. In October 1887 Duhem was appointed Maître de Conférences (lecturer) at the Faculty of Sciences of the University of Lille, stronghold of ultraconservative French Catholicism. In Lille, at the age of 28, in 1890, he met and married Marie-Adèle Chayet. Their daughter, Hélène, was born in September 1891. Adèle died in childbirth the next summer. The newborn child also did not survive. Duhem never remarried. He left Hélène's upbringing to his mother, who lived with him after his father died. After a six-year stay in Lille and a promotion to chargé de cours (assistant professor), he moved for one year to the University in Rennes.

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[1] Whenever Duhem's daughter needed a little disciplining, her father showed her a picture of Berthelot as the one who would bring punishment [1].

Duhem was offered a university position in Bordeaux in October 1894. and his friends advised him to accept the position, saying improbably: “The road to Paris goes through Bordeaux” [5]. Duhem remained in Bordeaux until the end of his life, a little more than twenty years later. In 1895 a chair in physics was created for him. Eight well-known French scientists earned their doctorates in Bordeaux under Duhem: E. Monnet (calorimetric study of the dissociation of a salt), H. Pélabon (dissociation of hydroselenic acid), L. Marchis (displacement of the zero in thermometers), A. Turpain (Hertzian waves), F. Lenoble (permanent deformation of metal wires), P. Saurel (equilibrium of chemical systems), H. Chevalier (changes in electrical resistance caused by temperature), and F. Caubet (liquefaction of gas mixtures). While at Bordeaux, Duhem published many books, among them his four-volume treatise on chemical mechanics (1897–1899), his 500-page textbook of chemistry and thermodynamics (1903), which was promptly translated into English under the title *Thermodynamics and Chemistry: A Non-mathematical Treatise for Chemists and Students of Chemistry*, and masterpieces such as *La Théorie Physique, son Objet et sa Structure*, and *Essai sur la Notion de Théorie Physique.* In 1913 he began publication of *Le Système du Monde*, *Histoire des Doctrines Cosmologiques, de Platon à Copernic* (1913–1917), but only five of the intended ten volumes were written before his death. The prodigious quantity (more than 450) and quality of Duhem’s publications in many fields of science, the philosophy of science, and the history of science did not change the judgment of exile from Paris. Duhem was sending so many publications per year to *Comptes Rendu* that the editor wrote him that without arguing the quality of the communications their frequency was exceeding the limits assigned by the journal, and it was necessary that the publications be spaced out over several weeks. Very late in life he was approached about a newly created chair in the history of science at the Collège de France in Paris, but he refused it. The proud and stubborn Duhem told his daughter: “I am a theoretical physicist. Either I will teach theoretical physics at Paris or else I will not go there” [2].

In his own eyes, Duhem was primarily a physicist. Like Ernst Mach, Wilhelm Ostwald, and J. Willard Gibbs, he championed the position called “energetics” or “energetism,” against the main scientific trends of his times, believing that generalized thermodynamics provided the foundation of all of physics and chemistry. Thus he was anti-Maxwellian, anti-atomist and ultimately, anti-relativist [5, 6]. An area most suited to the method of energetics was chemical mechanics. In many works Duhem tried to organize fully the entire field by taking his lead from Gibbs’ memoir *On the Equilibrium of Heterogeneous Substances*. The consequence that no unstable chemical equilibrium was possible at a constant temperature illustrated for Duhem a principal claim of energetics, namely, the absence of a rigid line of demarcation between physics and chemical mechanics.

Duhem spent his entire scientific life advancing energetism from his failed dissertation in physics to his mature treatise *Traité de l’énergétique* (1911). His work in the history and philosophy of science can be viewed as an attempt to defend the aims and methods of energetics [5] against the arguments of atomism[2] by providing what he thought were its incongruities and inconsistencies. He claimed that physical theories should be not be looked upon as ultimate explanations but only as representations of reality [1, 5]. He was particularly critical of the use of models in physical science. According to Duhem, model building is an occupation pursued by scientists with ample but shallow minds, a trait of English scientists (like Maxwell and Faraday!) [7]. In a famous reply to a comment that his physics was rooted in his religious beliefs, Duhem argued that it was rooted in the exigencies of teaching.

Many French and foreign institutions honored Duhem. He was awarded the honorary degrees of Doctor of Philosophy (Jagellone University of Cracow) and Doctor of Physics (Catholic University of Louvain); he was appointed honorary member of the Société Scientifique de Bruxelles; foreign associate member of the Academy Royal of Belgium; member of the Polish Academy of Sciences; corresponding member of the Dutch Society of Experimental Physics and of the Reale Istituto Veneto di Scienze, Lettere, e Arte; honorary associate of the Reale Accademia dei Scienze of Padua; and full member of the Académie des Sciences in 1913. On the occasion of his appointment to the Académie, the daily *Figaro* wrote (December 9, 1913): “The new academician will be one of the youth in the Institut. He is hardly fifty. His works have borne on mathematical physics and his initial effort was a master stroke, because his first book was that *Potentiel Thermodynamique* where he showed chemists a road to follow and signaled a method whose direction proved its fruitfulness. He studied from the mathematical point of view almost all branches of physics. Duhem defends his own ideas with verve. He belongs to the race of fighting scientists. At any rate, those disputes, lively as they are, cannot but be profitable for science. It is from the shock of ideas that light has always sprung [1].”

Duhem was also awarded some prestigious prizes: Cross of the Legion d’Honneur, Prize Binoux (Académie des Sciences, 1909), and twice the Petit d’Ormoy Prize (Académie des Sciences, 1907 and 1919; the second was posthumous). In Bordeaux there is a street named Rue Pierre Duhem.

Duhem died of heart problems at the age of 56 on September 4, 1916 in Cabrespine. The obituary, written by Prof. P. Cousin in the *Rapports* of the University of Bordeaux, vividly summarizes his life: “Duhem’s first scientific memoir was an act of scientific independence. He never bowed before the sole authority of a name, however illustrious. He claimed the right to have his views discussed and always used that right with scientific honesty, having no other aim that the unfolding of truth” [8].

Duhem and Thermodynamics

Duhem was a very prolific writer; he wrote almost 450 papers and books in the different areas in which he was active. We will discuss briefly the ones in the area of thermodynamics that survived the test of time and led to equations that carry his name. All these equations are quoted in most text books in thermodynamics.

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[2] Atomism is a belief system that holds that by describing the particle composition of material, an explanation is thereby produced not only of the universal physicality but of reality. It would reduce thought itself to contingent atomic reactions.

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The Gibbs Phase Rule and Duhem's Theorem. Assume a multicomponent and multiphase system in equilibrium at a certain temperature and pressure. The Gibbs phase rule states that if no chemical reactions occur then the number of degrees of freedom L is given by the relation

$$L = C + 2 - F \tag{1}$$

where C is the number of components and F the number of phases present. The statement *degrees of freedom* expresses the number of *intensive variables* that can be fixed arbitrarily and still retain the original number of phases of the system. Analysis of the phase rule indicates that once the intensive state of a system is established, all of its intensive properties are fixed, including the molar or specific properties of the phases and the partial properties of all species in each phase. However, nothing is implied about the *relative* amounts of the phases. A particularly interesting case of the Gibbs rule analyzed by Duhem is that of closed systems (constant mass), for which the extensive *and* the intensive state of the system are fixed. The states of such systems are characterized not only by the $[2 + F(C - 1)]$ *intensive* coordinates required by the phase rule but also by the F *extensive* coordinates represented by the number of moles in each phase. Therefore the total number of variables is $2 + F(C - 1) + F = 2 + FC$. If the system is closed and no chemical reactions are taking place, then the different compounds can transfer from one phase to the other, but the total number of moles remains constant. In other words, we can write a material balance equation for each of the C chemical species and the total number of independent equations becomes $C(F - 1) + FC = CF$. The difference between the total number of coordinates and the number of independent equations relating them is $2 + CF - CF = 2$.

This remarkable result is the basis of *Duhem's theorem* [9], which states that for any closed system formed initially from given masses of prescribed chemical species, the equilibrium state is completely determined by *any two* independent coordinates of the system.

The two independent variables that are to be specified may, in general, be intensive or extensive. However, the number of intensive variables is that given by the phase rule. This means, for example, that at the triple point of a pure component where $L = 0$, both variables required by Duhem's theorem must be extensive. For the vapor–liquid equilibrium of a pure component were $F = 1$, at least one of the variables must be extensive.

What happens to Duhem's theorem if chemical reactions are present? Actually, nothing. Each chemical reaction determines a new variable (the conversion) and a new coupling (stoichiometry) between the components. Therefore the difference between the number of variables and the number of equations remains unchanged, and Duhem's theorem also remains unchanged.

Gibbs–Duhem Equation

Consider a multicomponent system in a state of equilibrium at a certain value of the pressure and temperature. Any extensive property of the system, M, will be a function of the pressure, temperature, and number of moles of the different components, that is, $M = M(P, T, n_1, n_2, \ldots\ldots n_i)$. Taking the total differential of M yields

$$dM = \left(\frac{\partial M}{\partial P}\right)_{T,n_i} dP + \left(\frac{\partial M}{\partial T}\right)_{P,n_i} dT + \sum_i \left(\frac{\partial M}{\partial n_i}\right)_{P,T,n_j} dn_i \tag{2}$$

Subscript n_j means that the number of moles of all components different from component i are held constant. Since, by definition

$$\left(\frac{\partial M}{\partial n_i}\right)_{P,T,n_j} = \bar{M}_i \tag{3}$$

where $\bar{M}_i$ is the partial property, then

$$dM = \left(\frac{\partial M}{\partial P}\right)_{T,n_i} dP + \left(\frac{\partial M}{\partial T}\right)_{P,n_i} dT + \sum_i \bar{M}_i dn_i \tag{4}$$

But $M = \sum_i \overline{M_i n_i}$ so that

$$dM = \sum_i \overline{M_i} dn_i + \sum_i n_i d\bar{M}_i \tag{5}$$

Comparing eqs. (4) and (5) gives finally

$$\left(\frac{\partial M}{\partial P}\right)_{T,n_i} dP + \left(\frac{\partial M}{\partial T}\right)_{P,n_i} dT - \sum_i n_i d\overline{M_i} = 0 \tag{6}$$

Equation (6) is the most general form of the *Gibbs–Duhem equation* [10] valid for any molar thermodynamic property M in a homogeneous phase. For example, if M is the enthalpy, then we have [11]

$$\left(\frac{\partial H}{\partial T}\right)_P = c_P$$

$$\left(\frac{\partial H}{\partial P}\right)_T = v - \left(\frac{\partial v}{\partial T}\right)_P$$

so that

$$\left[v - \left(\frac{\partial v}{\partial T}\right)_P\right] dP + c_P dT - \sum_i n_i d\bar{H}_i = 0 \tag{7}$$

The Gibbs–Duhem equation is one of the most important relations of classical solution thermodynamics, for it constrains the allowable composition dependencies of the partial properties, This is most readily seen by examination of the constant-T, P form of eq. (6)

$$\sum_i n_i d\bar{M}_i = 0 \tag{8}$$

Equation (8) indicates that the changes in the values of the partial properties are not independent; the equation adds an additional coupling among them, that is, it reduces the number of degrees of freedom by one. To understand the meaning of this fact, consider a binary solution

$$n_1 d\bar{M}_1 + n_2 d\bar{M}_2 = 0 \tag{9}$$

$$d\bar{M}_1 = -\frac{n_2}{n_1} d\bar{M}_2 \tag{10}$$

In other words, knowledge of the first partial property determines completely the value of the second.

The Gibbs–Duhem equation is the basis of most of the so-called tests for *thermodynamic consistency*. What do we mean by this concept? The equation gives us a tool to test for the quality of data. If we have two independent ways of determining how each partial property varies with the composition, then both determinations must satisfy the Gibbs–Duhem equation to be thermodynamically correct.

Duhem was the first to use Euler's theorem for homogeneous functions to derive equation $M = \sum \bar{M}_i n_i$, and from there eq. (6). As we have mentioned above, the derivation appears for the first time in 1886 as part of his first (failed) doctoral thesis. Duhem repeated the derivation in a four-volume set of books published in 1897–1899 [12].

The Duhem-Margules Equation. The Gibbs–Duhem equation can be written in terms of the activity coefficient γ, as follows. Assume that $M = G^E$, the excess Gibbs function, then

$$\left(\frac{\partial G^E}{\partial P}\right)_{T,n_i} dP + \left(\frac{\partial G^E}{\partial T}\right)_{P,n_i} dT - \sum_i n_i d\bar{G}_i^E = 0$$

but, by definition

$$\bar{G}_i^E = RT \ln \gamma_i$$

so that at constant pressure and temperature we have, after division by the total number of moles n

$$\sum_i x_i d \ln \gamma_i = 0 \tag{11}$$

or

$$\sum_i x_i \left(\frac{\partial \ln \gamma_i}{\partial x_i}\right)_{P,T} = 0 \tag{12}$$

Assume now a binary system in vapor–liquid equilibrium at a given pressure and temperature. Furthermore, let us assume that the vapor phase behaves like an ideal gas. For these conditions we can calculate the partial pressure of each of the components using the modified Raoult's law [11]

$$P_i = \gamma_i x_i P_i^0 \tag{13}$$

where P_i^0 is the vapor pressure of component i at the system temperature. Substitution of eq. (12) in eq. (13) yields

$$x_1 \left(\frac{\partial \ln P_1}{\partial x_1}\right)_{P,T} + x_2 \left(\frac{\partial \ln P_2}{\partial x_1}\right)_{P,T} = 0 \tag{14}$$

Equation (14), called the *Duhem–Margules equation* [13], is valid no matter how strong the liquid phase deviates from ideal behavior, and it depends only on the assumptions that the gas phase is ideal and that the total pressure is not extremely high to make the vapor pressure of the pure compounds dependent on it.

Combining eq. (14) with the condition $P = P_1 + P_2$ and eliminating P_2 yields

$$\left[\frac{\partial P_1}{\partial x2} = \frac{(1 - x_1)P_1}{P_1 - x_1 P} \frac{\partial P}{\partial x_2}\right]_T \tag{15}$$

These equations enable us to calculate the partial pressures and the activity coefficients when we know the total pressure as a function of the composition. This is an interesting possibility since it is much easier to measure total pressures than partial pressures.

Conclusion

Pierre Duhem was a multifaceted and prolific scientist active in thermodynamics, physics, history, and philosophy. His rigid and noncompromising attitude, together with deep religious feelings, led to his being unable to teach in Paris and to academic exile in the provinces. He was a prolific writer and left his name on many equations, such as the Gibbs–Duhem and Duhem–Margules equations, that have survived the test of time

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