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PETER JOSSEPH WILHELM DEBYE

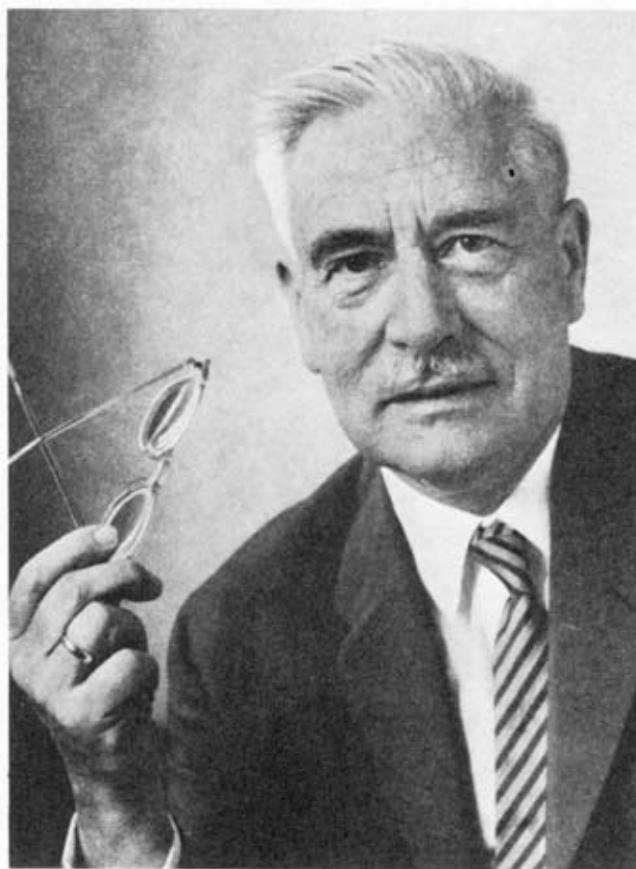
1884—1966

A Biographical Memoir by
J. W. WILLIAMS

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Biographical Memoir

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P. Debye

PETER JOSEPH WILHELM DEBYE

March 24, 1884–November 2, 1966

BY J. W. WILLIAMS

VITAE

PETER JOSEPH WILHELM DEBYE was born on March 24, 1884, at Maastricht, the Netherlands. His education began in the elementary and secondary schools there; it continued at the Technische Hochschule in Aachen. His first degree, achieved in 1905, was in electrical engineering. During the Aachen period Debye came under the influence of two exceptionally able physicists, Professors Max Wien and Arnold Sommerfeld, and with their encouragement and guidance remained there for a short additional period with an appointment as Assistant in Technical Mechanics. When Sommerfeld was called to Munich in 1906 as Professor of Theoretical Physics he invited Debye to accompany him as his assistant. Debye there completed his doctoral program in July 1908 and was promoted to privatdozent in 1910. By this time it was abundantly evident that he was well on the way to an illustrious career in physics.

In 1911 Debye received an appointment at the University of Zurich as Professor of Theoretical Physics. He returned to the Netherlands in 1912 to accept a position as Professor of Theoretical Physics at the University of Utrecht. The next invitation, two years later, was from Göttingen to take charge of the theoretical section of the Physics Institute. Within a

short time he became director of the institute, and he lectured on experimental physics until after the end of World War I.

Debye returned to Zurich in 1920, this time to become Professor of Physics and director of the Physics Laboratory at the Eidgenössische Technische Hochschule. An equivalent position at the University of Leipzig opened in 1927, and he was invited to fill it. From 1934 to 1940 he served as director of the Max Planck Institute of the Kaiser Wilhelm Institute for Physics at Berlin-Dahlem and Professor of Physics at the University of Berlin.

The Berlin post turned out to be his last in Europe. Immediately following its termination (for political reasons) he became Professor of Chemistry and, later, also chairman of the Department of Chemistry at Cornell University at Ithaca, New York. The promotion to emeritus status came in 1950. It was during the Ithaca period that Debye became an American citizen.

The quality of his scientific work gained him many honors and distinctions. A number of them have been listed to form an endpaper for this Memoir. Election to the National Academy of Sciences (U.S.) came first as a Foreign Associate (1931) and then as a Member (1947).

A different type of recognition came in 1939. A shoulder-length bust, a gift of the natives of his birthplace, Maastricht, was there unveiled in his honor to adorn the town hall. It has been noted by others that this distinction probably pleased Debye above all others.

Professor Debye married Mathilda Alberer in 1913. There were two children, a son, Peter Paul Rupprecht (b. 1916), and a daughter, Mathilda-Maria (b. 1921). He died on November 2, 1966.

THE SCIENTIST

In the Nobel Prize citation to Debye (1936) one reads, "for his contributions to our knowledge of molecular structure

through his investigations on dipole moments and on the diffraction of x-rays and electrons in gases." The structure of atoms and molecules was indeed a subject of major and continuing interest with Debye; it extended over the years from studies of the arrangements of the electrons in the simplest of the atoms to measurements of the average end-to-end distance in macromolecules of the "random-coil" type. One might have elected to consider in a single section those contributions that are related to the structure of matter, but here the attempt will be made to conform more closely to the outline Debye himself selected for his *Collected Papers* (1954), a volume that was presented to him by his students and friends and by the publisher on the occasion of his seventieth birthday, in 1954. In this way there is retained to some degree a chronological order, another plan that might have been adopted.

The articles presented in this compendium, fifty-one titles, constitute somewhat less than one quarter of the total number of his contributed papers. In even this portion one finds an impressive record of high-level achievement. The main subject areas are four in number: "X-Ray Scattering," "Dipole Moments," "Electrolytes," and "Light Scattering." A fifth unit is made up of "Miscellaneous Contributions." In the development and description of the researches, the reports are invariably replete with that same skill for which the author came to be known as a speaker and lecturer, namely, consummate proficiency in the description of a difficult and intricate subject in a lucid and well-organized fashion. A study of these and the other Debye contributions is indeed a rewarding experience.

X-RAY SCATTERING

The story has often been told of how, after learning about the progress of a study of the passage of light through crystals by Ewald and Sommerfeld at Munich in 1910-1912, Von Laue became interested in the passage of very short waves through such materials. He reasoned that if the wavelength of the

radiation were of the same order as the distance between the structural units a diffraction effect should be obtained. For the experimental test he suggested that x rays be used; the result strongly supported the correctness of his anticipation of a diffraction of the x rays by the crystals. As a result of this experiment a whole new subject, x-ray analysis, had been created.

Though the analysis came to be recognized as being simple in principle, there were certain complications in detailed application. Debye, well informed about the research activities at Munich, was quick to perceive that refinements of several kinds were necessary if the analyses were to have quantitative character. His treatments of two of them, the temperature effect (1914) and the atomic scattering factor effect (1915), are representative of great pioneering achievement.

In the first of these efforts Debye made calculations of the influence of the thermal vibrations of solids on the x-ray diffraction pattern. His earlier experience with the famous theoretical evaluation of the heat capacities of crystalline solids (1912) served him well in this endeavor. Using the same general idea, that the thermally induced atomic displacements in the crystal may be described as being elastic waves that are propagated through the material, he developed a mathematical expression to describe the temperature dependence of the x-ray structure amplitude factor. Introduced was the quantity now known as the Debye, or Debye-Waller, temperature factor. (Waller, later on, had made some adjustments.) Incidentally, this factor is essential to an understanding of the Mössbauer effect.

A consideration of the atomic scattering factor, Debye's second of the two refinements discussed in x-ray analysis, is of vital importance in structure determination. For the analysis, observed intensities of the spectra are compared with those calculated for assumed electronic arrangements of the structural elements. The calculations require a knowledge of the atomic scattering factor, a quantity that describes the result of interference effects within the scattering atoms.

For atoms of different sizes and kinds the scattering power for the x rays varies. Further, the waves scattered from the different parts of the electron cloud that surrounds a nucleus will be diffracted with phase differences in the direction of observation. The total amplitude is thus a function of the scattering angle and the distribution of the electron density about the atom. The atomic scattering factor, the quantity calculated, is defined as the ratio of the actual amplitude to that which would be produced by a single Thomson electron under the same experimental conditions.

Debye was able to take these several factors into quantitative account (1915). He demonstrated that as the angle of scattering increases, these phase differences become larger, so that the effective number of scattering units becomes smaller. The scattering factor, f , is now a quantity smaller than the total number of electrons in the atom. The factor depends on the wavelength, λ , of the incident rays in such a way that it is a function of $\sin \theta/\lambda$, with θ being the Bragg angle of diffraction. For example, it was possible for Debye to construct the curve for the distribution of diffracted x-ray intensity to be expected from Bohr atoms with their electrons arranged in circular orbits about their nuclei.

It was at this time, 1915, that Debye first recorded his conclusions that in matter of any state one never finds a completely random arrangement of atoms and molecules, and that perfect crystallinity is not required for the diffraction of x rays. It was pointed out in this renowned article (1915) that even in gases *the atoms are not completely random in their order*. This observation was the beginning of a whole sequence of experimental researches by Debye concerned with the scattering of x rays by gases, liquids, and amorphous solids. In such systems the curve of diffraction intensity versus angle of diffraction should show broad maxima and minima. However, Debye's first experimental test, conducted with Scherrer (1916), produced an unanticipated result. The test substance was finely powdered lithium fluoride,

but the x-ray diffraction pattern that was observed consisted of the sharp spots characteristic of diffraction by a crystal lattice. The formation of the sharp rings was properly explained as being due to the intersection on the photographic plate of a succession of conical beams from randomly oriented crystals. It was in this way that a new and useful method of x-ray analysis, the "powder method," was discovered.

Debye persisted in his researches in x-ray optics. In an article published somewhat later (1925), his thoughts were refined, extended, and summarized. He reaffirmed that it should be possible to observe diffraction effects that are interpretable in terms of the structure of the atoms and the molecules, irrespective of their physical state. More definitively, the thought was still to the effect that certain arrangements of any given atom with respect to its neighbors are more probable than others; thus it should be possible to obtain information about them by an x-ray analysis, regardless of the state of matter. For liquids on exposure to x rays a small number of broad and diffuse halos are produced in scattering. Two factors determine the outline of these halos; Debye early called them "inner" and "outer" interferences, with those of the first kind being between waves scattered by atoms belonging to the same molecule, while those of the second kind derive from intermolecular interferences. It is now known that this distinction cannot generally be made.

For the molecular structure determination it was reasoned that the "outer" interferences should vanish if the system were "diluted," as in a gas. In this way, the mathematical analysis and interpretation should be greatly simplified. The intensity factor, I , scattered by the gas should be an average effect, one described by a well-known Debye formula

$$I = k \sum_1^n \sum_1^n f_i f_j \frac{\sin x_{ij}}{x_{ij}},$$

in which the magnitude x_{ij} is proportional to the distance l_{ij} from atom i to atom j and f_i and f_j are their atomic scattering factors. The sums include the cases where $i = j$. For the angle of scattering, 2θ , of rays of wavelength λ (of the primary radiation),

$$x_{ij} = 4\pi l_{ij} \frac{\sin\theta}{\lambda}.$$

The formula is written for a molecule that consists of n atoms.

The scattering curve is thus a composite of as many individual curves as there are atomic distances in the molecule. Each such distance produces an intensity that increases and decreases as the angle of scattering is increased; the importance of the several interatomic distances is measured by the product of the scattering factors.

The results of the first experiments with gases, those from Debye's laboratory, were reported from Leipzig (1929). The reasoning had been correct; interference rings were produced by the scattering of x rays even from the simplest of molecules. For instance, from the photometered records of the rings, the chlorine-chlorine interatomic distances in carbon tetrachloride could be established with precision. Since the model for this molecule is taken to be a tetrahedron, this single distance suffices to define the complete structure. A more definitive and expanded account of similar researches, extended to certain other molecules, appeared within another year (1930).

It was at this time that Mark and Wierl* presented a preliminary description of their investigations showing that the Debye formula descriptive of the scattering of x rays by a gas, a brief outline of which appears above, could also be applied to describe the scattering of electron rays by gases. Physically, there is one difference. The electron interferences provide information about the positions of the atomic nuclei themselves, while the x-ray interferences reveal the locations of the centers

* H. Mark and R. Wierl, "Electron Diffraction, by a Single Molecule," *Naturwissenschaften* 18 (1930) : 205.

of gravity of the electron clouds about them. What is really ascertained in either case is the position of the atom centers, the desired quantity.

For reasons that need not be here described, electron diffraction became at once the preferred experiment. Though it is true that the actual number of molecules to which these methods may be applied remains small, still with modern computational devices and vastly improved equipment, electron diffraction has become a method of great utility and high precision for the evaluation of molecular structure.

Concurrently with the study of gaseous structure Debye, with Menke (1930), conducted experimental researches to determine the inner structure of liquids by x-ray analysis. The scattering pattern now represents the superposition of the two interference phenomena, an intramolecular part and an intermolecular part. It was argued that if these two parts could be separated, it would become possible to draw conclusions about the structure of the liquid. Mercury, a monatomic liquid, was selected as being a suitable test substance. With this choice the separation of inner and outer effects becomes possible. The separation of the contributions to the scattering pattern was achieved, and, by using an analysis of the type that had been presented already, by Zernicke and Prins,* it was possible to compute a distribution function to describe the probability of finding the molecules in the liquid at particular separations. This probability distribution curve for mercury demonstrated that even in the liquid there is found to be a quasi-crystalline state. The term "clustering" has been applied to short-range order situations of this general type; one finds it used in several other Debye discussions, in particular in his description of the underlying principles of electrolytic solution behaviors and in his treatments of the critical state.

* F. Zernike and J. A. Prins, "The Bending of X-Rays in Liquids as an Effect of Molecular Arrangement," *Zeitschrift für Physik* 41 (1927): 184.

In these ways it was proven that there is no absolutely sharp distinction between the amorphous and crystalline states. The general subject was again given definitive overall and more modern consideration in a critical review published rather recently (1960). The volume in which this article appears provides a good idea of the enormous amount of work that has been done in structural studies of various types of amorphous materials, an area in which Debye was the pioneer.

DIPOLE MOMENTS

It is as a consequence of their asymmetrical (electrical) structure that most molecules possess a permanent dipole moment; the magnitude of this characteristic entity is a quantitative measure of the polarity of the molecule. The practical unit of dipole moment is 1×10^{-18} e.s.u., now universally known as the debye, with symbol (D).

Sixty years after the appearance of the original Debye contributions on the subject (1912; 1913), the measurement and interpretation of molecular dipole moments continues unabated. Of the two articles, one was addressed to the problem of the behavior of a dielectric in a static electric field and the other to the case in which the electric field varies sinusoidally with time. The full significance of their teachings was not immediately recognized in chemical circles. The reports had appeared in journals for subjects in physics, and they were mathematical in character. This situation changed with the appearance of two more lengthy discourses by Debye: the renowned article in the Marx *Handbuch der Radiologie* (1925) and the record of a course of lectures presented at the University of Wisconsin in early 1927 and published later in book form (1929) with the title "Polar Molecules." In these publications the subject matter was superbly summarized, organized, and enriched, and it came quickly to the attention of the physicists, who in turn communicated their interest to friends

in chemistry. In the United States Professors K. T. Compton and R. C. Tolman were of great influence in this way.

To provide an indication of continuing interest we may note that beginning in 1955 at least three monographs that summarize advances in the subject have made their appearance: C. P. Smyth, "Dielectric Behavior and Structure," United States; N. E. Hill, W. E. Vaughan, A. H. Price, and M. Davies, "Dielectric Properties and Molecular Behavior," Great Britain; and V. I. Minkin, O. A. Osipov, and Yu. A. Zhdanov, "Dipole Moments in Organic Chemistry," the Soviet Union. An earlier Methuen pocket-size monograph, *Dipole Moments*, by R. J. W. LeFevre is now in its third edition; we consider this source of information to be an excellent introduction to the subject.

Debye, in his treatment of the electrical case, made use of the Langevin statistical theory of orientation for the permanent magnetic moments of paramagnetic molecules. In doing so, he took cognizance of the fact that matter is built up of electrically charged units. Prior to 1912 it had been recognized that many molecules, ammonia and water for example, showed abnormally high electrical susceptibilities, ones for which there was no explanation. By analogy with the magnetic problem Debye reasoned that such asymmetric molecules must possess finite and permanent electrical moments and that their total electrical polarizations result from two contributions, a displacement of electrons and atoms in the molecule and an orientation in the electrical field of the molecule as a whole. For the actual application in any given case it was necessary to devise means for the quantitative evaluations of each of these polarizations. The result, another well-known Debye equation that can be applied to polar gases at low pressures or (less exactly) to dilute solutions of polar substances in a nonpolar solvent, provides the means to compute the dipole moment, μ . It is, in molar form,

$$P = \frac{4\pi}{3} N (a + \mu^2/3kT).$$

The quantity P , which Debye called the molar polarization, is

evaluated experimentally by means of the Clausius–Mossotti formula, which involves dielectric constant and density data. As the formula is written, the term a measures the sum effect of electronic and atomic polarizations of a molecule as the field is applied; it is a constant that is independent of temperature. The quantity $\mu^2/3kT$ represents the orientation polarization, again per molecule. The equation demonstrates that a plot of P versus $1/T$ (T = absolute temperature) should be linear. From the slope of the line the dipole moment of the molecule is calculable.

At the time of its inception the dipole moment was the principal source of information about molecular structure. Now such data for small molecules of the rigid type have become of lesser significance because of the incidence of the x-ray and electron diffraction techniques (of Debye) and of modern spectroscopic methods.

There are molecules for which the P versus $1/T$ plot is nonlinear, with downward concavity. This result indicates that the molecular dipole moment is not independent of temperature; it can be explained by an intramolecular rotation of polar groups. Such effects are observed all the way from relatively small molecules to “random-coil” polymers in which, for example, -C-C- linkages occur. The particular finite value of the dipole moment observed at any temperature for molecules with internal rotations about such linkages then becomes the root-mean square value averaged over all the rotational positions.

For the molecule 1, 2-dichloroethane, for example, a rotation of the two -CH₂Cl groups about the -C-C-bond could be established. In this instance the rotations are of the “hindered” rather than “free” type. The molecular configuration possesses a center of symmetry when the two Cl atoms are at their greatest distance apart, the *trans* form, and the dipole moment in this arrangement is zero. In any other configuration, the dipole moment is finite and increases with increasing angle of internal

rotation; it reaches a maximum value in the *cis* form, with an azimuthal angle of 180° . A third rotational state, the *gauche* form, has an azimuthal angle of $\Phi = \pm 120^\circ$. These three rotational states serve to represent and characterize the staggered forms of the molecule.* In this way it becomes possible to compute a fractional occupation of each state and its relative potential energy.†

With F. Bueche, Debye (1951) applied this relatively simple idea of internal rotations to an organic high-polymer system. It will be indicated later that one may gain information about the average size of coiled polymer molecules in solution from light scattering measurements. However, the average coil diameter found by this experiment is usually much larger than would be calculated for a "random-coil" molecule of the same molecular weight and with unhindered rotation about the bonds that link the monomeric structural units. The difference was ascribed to a restriction of the rotations about these bonds, and a model was devised by which its effect could be quantitatively taken into account.

It may be mentioned that there have been established by dipole moment studies cases where free and unhindered rotations are encountered. An interesting example in polymer chemistry is that of the omega-hydroxydecanoic acid esters.‡ The result, the interpretation of which is actually a triumph of the Debye dipole theory, provided early and ample justification for the universal use by polymer chemists of the "random-coil" model for their macromolecules.

The concept of the orientation of dipolar molecules in an

* S. Mizushima, *Structure of Molecules* (New York: Academic Press, Inc., 1954), p. 7.

† Cf. for example, C. L. Braun, W. H. Stockmayer, and R. A. Orwoll, "Dipole Moments of 1,2-Disubstituted Ethanes and Their Homologs. An Experiment for Physical Chemistry," *Journal of Chemical Education* 47 (1970): 287.

‡ W. B. Bridgman and J. W. Williams, "Polar Group Orientation in Linear Polymeric Molecules. The ω -Hydroxydecanoic Acids," *Journal of the American Chemical Society* 59 (1937): 1579; J. Wyman, Jr., "A Dielectric-Constant Study of ω -Hydroxydecanoic Acid Polymers," *Journal of the American Chemical Society* 60 (1938): 328.

electric field, this time an alternating one, was applied by Debye (1913) in the explanation of the behavior of the two dielectric constants, real and imaginary, that are to be observed. (Permittivity and loss factor are better terms when frequency dependence is involved.) The basic principle is that when the field is applied, or released, a finite time will be required for the molecules to come to their equilibrium orientation because there is a viscous resistance to these rotatory motions. The range of frequency over which the real dielectric constant is variable extends from the static field to one that oscillates so rapidly as not to provide for any rotational motion of the polar molecule at all; the theory thus describes a typical molecular relaxation process. The accompanying constant, called the time of relaxation, is made available from measurements of the frequency variation of either the real dielectric constant or the energy absorption for the system; in solutions this time constant may be related to molecular size and shape.

The arguments and equations presented in connection with the frequency dependence problem have perhaps been of greatest interest in electrical engineering. One difficulty with the Debye theory has been that, written in terms of molecular dimensions and the internal friction of the medium, it leads to equations of quite the same mathematical form as an alternative explanation of Wagner * that is founded upon inhomogeneity of substance without reference to any molecular mechanism. There is nothing vague about the Debye model, and one can readily appreciate its appeal to those who work to elucidate the molecular behavior of electrically insulating materials. The model's main fault may be that it is too definitive in character.

ELECTROLYTES

As physical chemistry was taught in the early 1920's one of its major subdivisions was a description of the electrochemical

* Cf. in H. Schering, *Isolierstoffe in der Elektrotechnik* (Berlin: Springer, 1924), p. 1.

behavior of electrolyte solutions. But in this particular area one encountered many perplexing situations; overall its consideration was not a satisfying experience for either teacher or student. Clearly, a new idea was needed, and while Debye may not have provided it he did achieve great success in transforming a new postulate into an effective and practical working tool.

In particular, there was a fundamental problem in that the simple laws of Arrhenius and van't Hoff, so successful in application for the study of the equilibrium and transport properties of weak electrolytes (organic acids and bases), failed utterly when applied to account for these same kinds of data for solutions of the strong electrolytes (salts, certain inorganic acids and bases). In the latter situations the starting point, a mass action law equilibrium, was clearly inconsistent with the results of extensive sets of experiments. The answer was found in the assumption that the strong electrolytes are completely dissociated when dissolved in water. This representation had been considered by others, notably by Bjerrum and Sutherland and, using it, Milner * had actually computed the osmotic coefficient (a quantity that is simply related to the activity coefficient) for the electrolytes. Certainly, it may be said that the Milner analysis is, in principle, a solution of the thermodynamic problem, but there remained substantial mathematical difficulties such that the result had to be expressed in graphical and impractical ways.

For the treatment of the equilibrium properties, another mathematical route was selected by Debye and Hückel (1923). The results were presented in quantitative expressions that could be adapted simply and directly to freezing-point depression and related data for dilute strong-electrolyte aqueous systems. In the classical theory the ions had been treated as independently active units. In the new analysis it was the

* S. R. Milner, "Virial of a Mixture of Ions," *The Philosophical Magazine* 23 (1912): 551; "The Effect of Interionic Forces on the Osmotic Pressure of Electrolytes," *The Philosophical Magazine* 25 (1913): 742.

electrostatic forces exerted between the ions that proved to be the basic causes of the observed nonideality of solution behavior—hence the term “interionic attraction theory.” Actually, for sufficiently dilute electrolyte solutions, it became possible for Debye and Hückel to calculate in advance of any experiment what would be the observed osmotic pressure (or freezing point lowering, etc.) for salts of different valence types at a given ionic strength (a function of the electrolyte concentration) in an aqueous solution. The restriction to dilute solution behaviors made easier an otherwise very involved mathematical problem.

Still with reference to the first of the Debye–Hückel papers and the freezing point depression problem, we amplify these remarks. The argument is based on the application of well-known laws of electrostatics and together with the Maxwell–Boltzmann statistics. The ions were considered to be spheres of the same diameter, with their charges spread out in spherical symmetry. The solvent was a medium of uniform dielectric constant, a quantity unchanged by the addition of the solute ions.

The ions in solution might be expected to be in random thermal motion. However, because of the charges they carry, there will be, as a time average, more ions of opposite sign than those of the same sign in a neighboring small element of volume about any individual ion upon which attention is focused. As a result there is a structure in the system, one which is neither completely regular nor completely random in character. Each ion is thus subject to an average net electrostatic attraction by all of the other ions, and a clustering results. The magnitude of this attraction is a function of the product of the charges of the ions and the mean distance between them (concentration of the solution). The potential energy of any arbitrary central ion in the solution is lower as compared to what the energy would have been if the ion had possessed zero charge. The magnitudes of ionic attractions and repulsions were described by Coulomb’s law, a fact that leads eventually to the

square root of concentration behaviors for both equilibrium and transport behaviors observed by experiment in dilute solutions of strong electrolytes.

In the theory it was the most probable distribution of an ionic atmosphere about a central ion that was first determined. Then, the average electrical potential of a given ion due to the presence of all the other ions was calculated. The calculation involved the combination of the Poisson differential equation, in which potential is related to the average electrical charge density, with the Boltzmann distribution theorem. It was the approximations here introduced to effect a simplification of the mathematical problem that have become the cause of much later comment. Certainly, they restrict the application of the theory largely to dilute aqueous solutions.

With a knowledge of this potential, the excess free energy due to the electrostatic interactions was computed. It is related to the several measures of solution nonideality, for instance, the osmotic coefficient from freezing-point depression data, and even more simply, the activity coefficient.

In the common usage of today it is the activity coefficient which is sought. Debye (1924) was prompt in his appreciation of its advantage in use over the osmotic coefficient; he recognized that his earlier presentation with Hückel (1923) could be greatly simplified if written in terms of the activity coefficient. It is the equivalent of this second derivation of the limiting thermodynamic law that is almost universally reproduced in the modern physical chemistry texts. The title of this article is, in translation, "Osmotic Equation of State and Activity of Dilute Strong Electrolytes." In the introduction to this report one finds the German equivalent of the sentences, "Besides, I have in the meantime come across some laws on the activity of strong electrolytes which G. N. Lewis discovered in a purely experimental way. I am glad to have this opportunity to emphasize the special importance of these fine investigations, the

more so since the laws of Lewis can be explained very easily by the proposed theory."

The appreciably more difficult transport problem of electrical conductance was the subject of the second of the basic Debye-Hückel theoretical treatments (1923). On the basis of the Arrhenius theory, the variation of the equivalent electrical conductance with electrolyte concentration was explained by the change in the relative number of the ions, the carriers of the current, as a function of concentration—a law of mass action effect. While this explanation remains correct to a good approximation for weak electrolytes, it could not account for the square root of concentration decrease in equivalent conductance with increasing concentration that had already been found experimentally by Kohlrausch and others for the strong electrolytes. Here, per equivalent of electrolyte, the number of carriers of electricity remains substantially constant (in dilute solution); it is the ion mobilities that decrease with increasing electrolyte concentration, again an effect of the interionic attractions. The discussion was now focused on two properties of the ionic atmosphere, a relaxation time effect and an electrophoretic effect. Although this time absolute values could not be computed, Debye and Hückel were able to demonstrate that for the limiting law the two progressive decreases in ion mobilities with increasing salt concentration are each proportional to the square root of the equivalent concentration. In their first treatment of the transport problem Debye and Hückel did not fully take into account the effect of the Brownian movement of the ions during the time of their displacement in the electrical field. The required modification was provided by Onsager, and the combined result is called today the Debye-Hückel-Onsager theory.

With the appearance of these papers there began a whole new era in the treatment of systems containing electrolytes. Debye himself continued to recognize new areas in the subject

and to treat them with his customary aplomb. Immediately following the publication of the fundamental disclosures he indicated (1923) at length that his simple limiting law for the activity coefficient of a strong electrolyte can be directly applied in the explanation of the change in solubility of a difficultly soluble salt caused by the addition to the solution of a salt without a common ion. The quantity $\log s/s_0$ versus square root of the ionic strength is linear, and $\log s/s_0$ is a direct measure of $-\log \gamma_{\pm}$, for the difficultly soluble salt. The quantities s and s_0 are the solubilities of the saturating salt in the presence and absence, respectively, of the added electrolyte, and γ_{\pm} is its mean activity coefficient.

In two papers on the "salting-out effect," Debye (1925; 1927) showed that the separation of organic solutes from saturated aqueous systems on salt addition is largely a consequence of the inhomogeneous electrical field produced by the localized charges carried by the ions. Again, these accounts do not take into consideration the fact of the presence of other forces, especially in that one is no longer dealing with dilute solutions.

In further connection with the transport problem Debye and Falkenhagen (1928) reasoned that because of the finite time of relaxation of the ionic atmosphere there must be a frequency dependence of the electrical conductivity for a strong electrolyte in solution. Further it was indicated that the Wien observation of a deviation from Ohm's law when high field strengths are applied in the measurement could be interrelated with the frequency dependence problem. Although the experiments are difficult to perform, the detailed prediction of the dependence of conductance on the frequency of the applied field was later verified by direct experiment, an establishment of the sufficiency of the Debye model to explain not only the conductance behavior, but also to provide treatments of other transport problems, such as diffusion and viscosity, again for dilute strong electrolyte solutions.

The Debye papers descriptive of his interionic attraction

theory have influenced profoundly the course of research in several related areas. We mention but two of them:

1. The velocity of ionic reactions is modified as salt is added to the system. These effects are not generally due to any direct action of the salt; they are due rather to the electrostatic forces between the ions as they influence the velocity constants for the reaction. The bearing of modern electrolyte theory on the reaction kinetics has been treated in an authoritative fashion by Bell.*

2. There has been much confusion in the literature of the protein physical chemist as data for typical equilibrium and transport experiments have been interpreted. The system used is traditionally that of water, protein, and salt, a three-component system, one which requires detailed mathematical interpretation. By the simple expedient of the addition of excess salt, called "supporting electrolyte," and attention to certain experimental details, the influence of the charge on the macroion is largely suppressed, and the problem is reduced in complexity to that encountered in the analysis of a two-component, neutral molecule system. Casassa and Eisenberg † have discussed this problem as it relates to osmotic pressure, light scattering, and sedimentation equilibrium in such systems.

LIGHT SCATTERING

The final period of Debye's life began in 1940 when he arrived at Cornell University. Debye now applied his talents to macromolecular and colloid chemistry and began at once to provide new ideas and ways for their study. Now, the central theme was again the interaction of radiation and matter. He recognized that just as the wavelength of x rays is comparable to the size of atoms in the crystal, so is the corresponding length of light rays of the same order as the dimensions of the polymer molecules and colloidal particles. As the result one finds in the

* R. P. Bell, *Acid-Base Catalysis* (Oxford: Clarendon Press, 1941).

† E. F. Casassa and H. Eisenberg, "Thermodynamic Analysis of Multi-Component Solutions," *Advances in Protein Chemistry* 19 (1964): 287.

literature theoretical analyses by which light-scattering experiments may be interpreted in terms of macromolecular size, radius of gyration, and even of the end-to-end distance of the macromolecular unit when it has the random-coil configuration. There are also significant extensions of our knowledge of the structure of colloidal particles and porous solids.

Of the two kinds of mathematical analysis for light scattering from solutions, the vibrating dipole theory of Hertz * and the density fluctuation theory of von Smoluchowski † and of Einstein, ‡ the latter is the more generally applicable.

A basic quantity is the turbidity, τ , described by an exponential law of common form:

$$I = I_0 e^{-\tau r}.$$

This statement has as a source the vibrating dipole theory. In it I and I_0 are the scattered and incident light intensities, respectively, and r is the distance between the scattering center and the point of observation. The turbidity is thus the extinction coefficient in cm^{-1} . In experimental quantities it can be written as:

$$\tau = \frac{32\pi^3}{3} \frac{\nu}{\lambda^4} \left[\frac{1}{n_0} \frac{dn}{dc} \right].$$

It will be noted that this formula contains the familiar Rayleigh scattering factor $(\lambda^4)^{-1}$. With ν , the number of macromolecules per cubic centimeter, which is equal to cN/M , the molecular weight (M) is introduced and $\tau = HMc$. The concentration (c) is the weight of solute per cubic centimeter of solution, H is the familiar light-scattering constant in $(\lambda^4)^{-1}$, and dn/dc is the refractive index increment.

Fluctuation theory may be used with advantage in the calculation of the excess scattering by a dissolved substance. From an

* H. Hertz, "Die Kräfte electrischen schwingungen, behandelt nach der Maxwellschen theorie," *Annalen der Physik* 36 (1888): 1.

† Smoluchowski, *Annalen der Physik* 25 (1908): 205.

‡ A. Einstein, "Theory of the Opalescence of Homogeneous and of Mixed Liquids in the Neighborhood of the Critical Region," *Annalen der Physik* 33 (1910): 1275.

Einstein relation between osmotic pressure and light scattering Debye obtained the general expression to describe the light-scattering behavior as a function of solute concentration (c) for a binary nonideal solution of neutral macromolecules whose size is small in comparison with the wavelength of the monochromatic light. It is:

$$\frac{Hc}{\tau} = \frac{1}{M} + 2Bc,$$

in which B is the osmotic pressure second virial coefficient.

With intramolecular interference of the light, such as is found in larger and flexible molecules, the situation becomes much more complicated because a particle-scattering factor now must be introduced into the essential working equations. Debye's classical and original work on the atomic scattering factor in x-ray analysis pointed the way for him to relate the angular dissymmetry of the light scattering now involved to particle shape. The particle-scattering factor contains a size parameter, angle of scattering, and wavelength dependence.

The application of the Einstein and Rayleigh equations for light scattering to the determination of the molecular size of macromolecules in solution did not originate with Debye. There were the earlier Putzeys and Brosteaux* and the Gehman and Field† contributions; proper and generous references to these papers were made in the Debye reports. But, as is typical, Debye did make the procedure a practical one, so much so that immediately following the appearance of his disclosures there began an explosive development of organic high-polymer chemistry—the subject matter of which had now been taken out of the realm of the descriptive and into exact science.

Further, the light-scattering techniques were applied to other types of systems, such as silicates and soap micelles. In an interesting series of papers, published during the period 1948–1951,

* P. Putzeys and J. Brosteaux, "The Scattering of Light in Protein Solutions," *Transactions of the Faraday Society* 31 (1935): 1314.

† S. D. Gehman and J. E. Field, "Colloidal Structure of Rubber in Solution," *Industrial and Engineering Chemistry* 29 (1937): 793.

Debye, with several collaborators, used the techniques to learn about micelle formation in solutions of paraffin-chain salts. Both the size and the shape of the micelles were considered. An objective was to determine the number of primary units of which they are composed and whether all the micelles are alike in size; another was to describe the mechanism of micelle formation.

Of the two theoretical accounts of the Debye theory of micelle formation appearing in the literature (1949), the New York Academy of Sciences article is the more definitive. It should be mentioned, however, that both Reich * and Ooshika † believe Debye to be seriously in error in his treatment of the problem. Both critics agree that the stable micellar size must be the one that results in a minimum of free energy for the system as a whole rather than for the individual micelle, as Debye had postulated. (Although an interval of two years between the appearance in the literature of the two criticisms is evident, the original manuscripts reached their respective editorial offices in August 1953, actually within one week of each other. The presumption is that they were conceived and published independently.)

In the final period of his scientific life, Debye became greatly interested in the phenomenon of critical opalescence and lectured widely and enthusiastically on the subject. Under certain circumstances small molecules may form aggregates of a size comparable to the wavelength of light, again a typical clustering phenomenon. A study of the scattering of light from such systems provides information about the distance of nearest approach of the molecules, which is taken to be a measure of the range of molecular interaction. But here, and apparently without full realization, the work of Debye had been largely, but

* I. Reich, "Factors Responsible for the Stability of Detergent Micelles," *Journal of Physical Chemistry* 60 (1956) : 257.

† Y. Ooshika, "Theory of Critical Micelle Concentration of Colloidal Electrolyte Solutions," *Journal of Colloidal Science* 9 (1954) : 254.

not completely, anticipated by Ornstein and Zernike*; their distribution function to ascertain this distance was the one used by Debye. Their study was concerned with density fluctuations in the critical region; the basic article bears the title "Accidental Deviations of Density and Opalescence at the Critical Point of a Single Substance." There exists in the literature a series of papers over a period of some eight years in which these authors continued the development and description of various phases of the problems related to the clustering tendency of molecules in the critical state and the resultant opalescence.

MISCELLANEOUS

Of the papers assigned to the miscellaneous category by Debye for his *Collected Works*, lack of space requires that but two of the items receive mention. Both are of such consequence that accounts of them appear in most of the better modern texts of physical chemistry.

The earlier one has to do with the theory of the heat capacity of solids (1912). According to an old empirical rule of Dulong and Petit, the heat capacity per gram-atom of an element in the solid state is 6.2 calories. In the attempt to account for this value theoreticians had believed it to result from an equipartition of energy, but as more accurate data for the temperature dependence of the heat capacity were made available it became evident that this could not be the complete explanation, especially at the lower temperatures. The experimental fact is that the heavy and soft elements possess this value for the heat capacity per gram-atom at room temperature, but for the light and hard elements much higher temperatures are required for its attainment. At the low temperatures all the solid elements show heat capacities lower than 6.2 calories per gram-atom.

* L. S. Ornstein and F. Zernicke, "Accidental Deviation of Density and Opalescence at the Critical Point of a Single Substance," *Proceedings of the Royal Academy of Sciences of Amsterdam* 17 (1914): 793.

The model used by Debye was to treat the solid as a continuum filled with elastic waves rather than as a system of oscillators. In spite of very involved mathematical operations he succeeded in deriving a formula that gives an excellent representation of the heat capacity at constant volume, c_v , as a function of temperature. It contains his famous T^3 law for the quantity c_v at very low temperatures, while still accounting for the fact that c_v does not increase indefinitely as the temperature is increased.

The later miscellaneous Debye paper, "Some Remarks on Magnetization at Low Temperatures" (1926), is another thoroughly imaginative and impressive item. (The same procedure was described independently by the American physical chemist Giauque, and concurrently in time.) Herein is presented in detail the principle of adiabatic demagnetization as a method for the production of very low temperatures. In it, a paramagnetic salt is inserted between the poles of a powerful magnet contained in a bath of liquid helium. As the field is turned on, the magnetic dipoles are oriented, with the production of heat; in turn, this heat is absorbed in the bath. Following the strong magnetization, the salt is insulated from its surroundings. On decrease of the field strength the orientation of the dipoles moves toward randomization, increasing the potential energy at the expense of the kinetic energy of the molecules. Accordingly, the temperature of the salt is decreased. The difficult experiment came gradually into fruitful application, beginning with Giauque in 1933.

Debye did not write many monographs and review articles. In addition to the definitive article on dipole theory in the Marx *Handbuch der Radiologie* (1925; 1934), his "Polar Molecules" of 1929 has served as a great stimulus to chemists. Interest in this volume has continued, with paperback reprints having been made available in 1945 and again in 1960. The Chu and the Prock-McConkey books, records of two series of lectures by

Debye (Cornell and Harvard), provide valuable information about molecular interactions and the forces responsible for them.

The overall record, of which but a small part has been herein depicted, must demonstrate that in his lifetime Professor Debye made many brilliant contributions of great value to physics, to chemistry, and to certain of their borderline disciplines. In these writings he has left a precious legacy for physical scientists. In every sense and by universal acclaim he was indeed one of the leading scientists of our century.

THE MAN

As an individual Professor Debye was held in universal affection and esteem by those who knew him. One description, taken from a Harvard University citation, is particularly apt—"a large-hearted physicist who gladly lends to the chemist a helping hand." He was the kind of person Maurice Hindus had in mind when he wrote, "A student needs to come under the influence of only one exciting professor to feel the effects of it all his life, even to have the course of his life changed." He was readily approachable, a very friendly person to whom one could go for advice in research and come away fully rewarded. No one was beneath his personal encouragement; he was patient and understanding with all.

The many honors and distinctions that came with the passing of the years did not in any way change him. He was modest and realistic about them. He never forgot his old friends and associates, nor did his interest in science diminish with increased time or fame. To the end his generosity, friendliness, and concern for others were commensurate with his mental prowess.

Whether as classroom teacher or as special lecturer he was renowned for his facility of expression. This apparent ease of exposition must have required concerted effort at organization. Nowhere were his abilities to explain scientific principles better

demonstrated than in his lectures for the large introductory physics courses presented during the Zurich and Leipzig periods. The concomitant lecture table displays were correspondingly pertinent and skillful; here again it was obvious that much thought and time had gone into their preparation.

In his years in the United States Debye became an inveterate traveler. He gave lectures and seminars outside of Ithaca almost weekly. At meetings his appearances invariably meant large audiences, for from his discussions at them the new and unexpected was the rule. He possessed the ability to explain scientific ideas and principles to a wide variety of audiences, and wherever he went he was received as a desirable and agreeable lecturer.

It has been noted elsewhere * that Debye was "an affectionate husband, father, and grandfather." His hobbies were few, such as gardening, fishing, and collecting cacti. There were periods when his lengthy activities in his rose garden might have brought concern to an observer, but more often than not they were followed by extraordinary bursts of scientific activity; a new idea had been elaborated during the out-of-doors time.

As a result of my own relationships with him I must note that Professor Debye did indeed have true kindness of heart, along with his rare vigor of intellect.

THE AUTHOR is indebted to colleagues both here at the University of Wisconsin and at the California Institute of Technology for their advice and help. Drs. E. W. Hughes, W. E. Vaughan, P. Bender, and J. D. Ferry have read portions of the manuscript, have given wise counsel, and have made useful suggestions.

Subject to certain revisions and modifications, the bibliography which is here included has been taken from the Debye Memoir written by Professor Mansel Davies for the 1970 volume, *Biographical Memoirs of the Fellows of the Royal Society* (London). Too, he has read the manuscript and raised certain questions in connection with it. For the permission of Dr. Davies to make use of both bibliography and suggestions, I am deeply grateful.

* F. A. Long, "Peter Debye—An Appreciation," *Science* 155 (1967): 979.

A substantial portion of this memoir was written in the hospitable Millikan Library of the California Institute of Technology at Pasadena.

HONORS AND DISTINCTIONS

ACADEMIES

National Academy of Sciences, Washington, D.C.
New York Academy of Sciences, New York
American Academy of Arts and Sciences, Boston
American Philosophical Society, Philadelphia
Franklin Institute, Philadelphia
Royal Dutch Academy, Amsterdam, Holland
Royal Society, London, England
Royal Institution of Great Britain, London, England
Royal Danish Academy, Copenhagen, Denmark
Academies of Berlin, Göttingen, Munich, Germany
Academies of Brussels and Liège, Belgium
Royal Irish Academy, Dublin, Ireland
Papal Academy, Rome, Italy
Indian Academy, Bungalow, India
National Institute of Science, India
Real Sociedad Española de Física y Química, Madrid, Spain

MEDALS

Rumford Medal (Royal Society, London), 1930
Lorentz Medal (Royal Dutch Academy, Amsterdam), 1935
Nobel Prize in Chemistry, 1936
Franklin Medal (Franklin Institute), 1937
Willard Gibbs Medal (American Chemical Society, Chicago), 1949
Max Planck Medal (German Physical Society), 1950
Nichols Medal (American Chemical Society, New York), 1961

HONORARY DEGREES

Brussels and Liège (Belgium)
Oxford (England)
Prague (Czechoslovakia)
Sofia (Bulgaria)
Aachen and Mainz (West Germany)

Zürich, E. T. H. (Switzerland)

Harvard, St. Lawrence, Colgate, Notre Dame, Holy Cross, Brooklyn Polytechnic, Boston College, Providence College, and Clarkson Institute of Technology (United States)

LECTURESHIPS

Paris (France)

Liège (Belgium)

Oxford and Cambridge (England)

Harvard, Michigan, Columbia, California, Southern California, Massachusetts Institute of Technology, California Institute of Technology, and Wisconsin (United States)

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KEY TO ABBREVIATIONS

- Angew. Chem. = Angewandte Chemie
 Ann. Phys. = Annalen der Physik
 Ber. Verh. Saech. Akad. Leipz. math.-naturwiss. Kl. = Berichte über die Verhandlungen der Saechsischen Akademie zu Leipzig, mathematisch-naturwissenschaftlich Klasse
 Bull. sci. Acad. Roy. Belg. = Bulletin des sciences, Academie Royale de Belgique
 C. R. Soc. Suisse Phys. = Comptes rendus de la Société Suisse de Physique
 Ergeb. tech. Röntgenk. = Ergebnisse der technischen Röntgenkunde
 J. Appl. Phys. = Journal of Applied Physics
 J. Chem. Phys. = Journal of Chemical Physics
 J. Colloid Sci. = Journal of Colloid Science
 J. Phys. Chem. = Journal of Physical Chemistry
 J. Phys. Colloid Chem. = Journal of Physical and Colloid Chemistry
 J. Polym. Sci. = Journal of Polymer Science
 Nachr. Akad. Wiss. Goett. math.-phys. Kl. IIa = Nachrichten der Akademie der Wissenschaften in Goettingen, mathematisch-physikalische Klasse, IIa
 Natur-en Geneesk. Congr. = Natuur-en Geneeskundige Congress
 Phys. Eindhoven = Physica, Eindhoven
 Phys. Rev. = Physical Review
 Phys. Z. = Physikalische Zeitschrift
 Rev. univ. mines (Liège) = Revue universelle des mines (Liège)
 Sitzungsber. Bayer. Akad. Wiss. math.-naturwiss. Kl. = Sitzungsberichte der Bayerischen Akademie der Wissenschaften, mathematisch-naturwissenschaftliche Klasse
 Trans. Am. Electrochem. Soc. = Transactions of the American Electrochemical Society
 Trans. Faraday Soc. = Transactions of the Faraday Society
 Verh. Dtsch. Phys. Ges. = Verhandlungen der Deutschen Gesellschaft für Physik
 Verh. Schweiz. Naturforsch. Ges. Freib. = Verhandlungen der Schweizerischen Naturforschenden Gesellschaft, Freiburg
 Z. Elektrochem. = Zeitschrift für Elektrochemie und angewandte physikalische Chemie
 Z. Phys. = Zeitschrift für Physik
 Z. phys. Chem. = Zeitschrift für physikalische Chemie
 Z. tech. Phys. = Zeitschrift für technische Physik

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