Lars Onsager (1903–1976), Chemist-Physicist, on the Silver Anniversary of His Death

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Abstract: The Norwegian-born theoretical chemist–physicist Lars Onsager (1903–1976) received the 1968 Nobel Prize for Chemistry "for the discovery of the reciprocal relations bearing his name, which are fundamental for the thermodynamics of irreversible processes." A recipient of numerous awards and honorary degrees, which he did not receive until relatively late in life, he taught at the Johns Hopkins and Brown u,niversities, but spent most of his academic career at Yale University (1933–1972). He spent his post-retirement years (1972–1976) as Distinguished University Professor at the University of Miami's Center for Theoretical Studies, where he continued his work with several postdoctoral research fellows.

October 5, 2001 marks the 25th anniversary of the death of Norwegian–American Nobel chemistry laureate Lars Onsager (Figure 1). In his Nobel presentation speech of December 10, 1968 Stig Claesson of the Royal Swedish Academy of Sciences told Onsager,

> You have made a number of contributions to physics and chemistry. For example, your equation for the conductivity of strong electrolytes; your famous solution of the Ising problem, making possible a theoretical treatment of phase changes; or your quantisation of vortices in liquid helium. However, your discovery of the reciprocal relations takes a special place. It represents one of the great advances in science during this century [1a].

Early Life

The son of Norwegian Supreme Court barrister Erling Onsager and teacher Ingrid Onsager (née Kirkeby), Lars Onsager was born on November 27, 1903 in Christiania (renamed Oslo in 1925), the capital of Norway [1c–12]. He was raised in Christiania, where he studied literature, philosophy, music, the fine arts, and the Norwegian epics. He spent three years with experienced educators Inga Platou and Anna Platou in Christiania and a year in a deteriorating private school in the country. After several months with his mother's tutoring when his family returned to Christiania, he entered the Frogner School, where he skipped a grade, enabling him to graduate in 1920.

Exhibiting an early interest in science and mathematics, in the fall of 1920 Onsager enrolled as a student of chemical engineering in the Norges Tekniske Høgskole (Norwegian Institute of Technology; since 1970, the University of Trondheim) in the port city of Trondheim about 230 miles north of Christiania. In this stimulating environment Professors O. O. Collenberg and B. Holtsmark (1859–1941) encouraged his theoretical work. Onsager solved most of the notoriously difficult problems in a calculus textbook [13] and began to read the original literature. In 1925 he received his chemical engineering (Ch.E.) degree and remained for an additional year (1925–1926) at Trondheim as a resident scientist.

As a student of 20, Onsager discovered a correction for Brownian motion to the recently published Debye–Hückel theory, which applied the Arrhenius theory of electrolytic dissociation [14, 15] to strong electrolytes [16]. He discovered that the electrostatic atmosphere produced by ions of opposite charge, which Debye and Hückel had postulated to shield each ion in solution, violated fundamental requirements of symmetry [1b].

In 1925, at age 22 and while still a student, Onsager traveled to Denmark and Germany. He then went to Zürich, Switzerland, where he spent several months at the Eidgenössische Technische Hochschule (ETH, Federal Institute of Technology) with Peter J. W. Debye (1884–1966), the future (1936) Nobel chemistry laureate [17], and his assistant Erich Hückel (1896–1980) [18]. Debye was so impressed with Onsager's correction to his theory that he offered him a research assistantship at the ETH for almost two years (1926–1928). With Debye's encouragement Onsager organized, developed, and published his results on the theory of strong electrolytes [19], which became known as the Onsager limiting law for dilute solutions of electrolytes. During his stay in Zürich he also met many prominent physicists and rowed in crew races on the Zürchersee (Lake Zürich).

Emigration to America

In 1928 Onsager accepted a position as a teaching associate at the Johns Hopkins University in Baltimore, Maryland, where he was assigned to teach freshman chemistry; however, his students complained that he did not present his lectures in suitably elementary terms. In fact, throughout his career the challenges presented by his lectures, even to experts, remained formidable. He habitually omitted intermediate steps in mathematical derivations. Preferring to overestimate his audience's abilities, he routinely assumed that they were as intelligent as he was. Not surprisingly, he was dismissed after only one semester, and he spent the spring 1929 semester formulating the work on reciprocal relations that was to win him the Nobel Prize for Chemistry almost four decades later.

Figure 1. Lars Onsager (1903–1976) (Courtesy, the Royal Swedish Academy of Sciences).

The Brown Years

In the fall of 1929 Chemistry Department Chairman Charles A. Kraus (1875–1967) [20] appointed Onsager a research instructor at Brown University in Providence, RI, where he remained for five years (1929–1933). Here he had no undergraduate teaching duties but taught a graduate course in statistical mechanics, which his students soon called "Advanced Norwegian I" or "Sadistical Mechanics," for his accent and obscure lecturing style made the difficult subject even more troublesome. He also began his more than three decades-long collaboration on electrolytes with his first coauthor, Raymond Matthew Fuoss, who was then a graduate student and later Onsager's fellow faculty member at Yale University. During his first year at Brown University Onsager submitted his work on reciprocal relations in simultaneous irreversible processes under nonequilibrium conditions as a Norwegian abstract for a meeting of the Scandinavian Physical Society at Copenhagen in 1929 [21]. It was destined to bring him the Nobel Prize for Chemistry—but not until 1968—almost four decades later.

While at Brown, Onsager sent Debye a manuscript for publication in the *Physikalische Zeitschrift,* of which Debye was editor, describing a correction to Debye's formula for the dielectric constant of polar molecules, but Debye did not approve. The so-called Onsager formula did not appear in print until John G. Kirkwood (1907–1959) [22] persuaded him to rewrite the manuscript in English and publish it in an American journal [23]. Kirkwood was an International Research Fellow with Debye at Leipzig and, like Fuoss, was destined to be a fellow colleague of Onsager's at Yale. Not until many years later did Debye finally accept Onsager's correction.

Kraus suggested that Onsager also carry out experimental research in addition to his theoretical analysis. The Soret effect—the development of a concentration gradient in a solution placed in a temperature gradient—is the type of "coupled flows" situation involved in Onsager's Reciprocal Relations and a practical application of abstract principles of the kinetic theory of gases to the separation of an isotopic gas mixture. With his five years of experience in chemical engineering, Onsager explored the separation of isotopes by thermal diffusion [24, 25], but his apparatus required the construction of a platinum tube three stories high—an impossible expense during the Great Depression, and he never completed the project. During World War II the separation of uranium isotopes, a crucial step in the production of the nuclear bomb, was accomplished by the gaseous diffusion of uranium(VI) fluoride as part of the Manhattan Project [26].

A Rapid Romance

Because of the Depression, in 1933 Onsager's position at Brown was eliminated, and he spent several weeks that summer in Europe with Hans Falkenhagen, the Austrian electrochemist of the Universität Köln (Cologne), with whom he had been corresponding. Falkenhagen was unwell and asked his sister Margarethe (Gretl) Arledter, the daughter of a famous pioneer in the art of paper making, to entertain him:

> Gretl saw him coming up the stairs—a very handsome young man who her brother had told her "was well ahead of his times." They went out to dinner, but Lars was his usual reticent self. After dinner he fell asleep on the patio, and then woke up and asked: "Are you romantically attached?" They became engaged 8 days later—Margarethe at 21 and Lars at 29—and got married [in Cologne] on 7 September 1933 [8].

The couple had a daughter, Inger Marie (Mrs. Kenneth Roy Oldham), and three sons, Erling Frederick, Hans Tanberg, and Christian Carl.

The Yale Years

In the fall of 1933 Onsager and his new bride returned to the United States where he accepted the prestigious Sterling postdoctoral fellowship (1933–1934), which did not involve any teaching duties, at Yale University. However, after his arrival, the Yale chemistry faculty was embarrassed to realize belatedly that Onsager did not possess a doctorate. They waived the course requirements and in 1935 awarded him a doctorate on the basis of a paper dealing with the mathematical background for his interpretation of deviations from Ohm's law in weak electrolytes. Because neither the Yale chemists nor physicists were able to evaluate it [27], (Carl) Einar Hille (1894–1980) of the Mathematics Department, an authority on the subject, read it and enthusiastically recommended it for a Ph.D. in mathematics. The Chemistry Department apparently awarded the degree themselves, for the problem had arisen in the chemical context of electrolytes [19, 28] and of ionized gases, in which Brownian motion must be taken into account. Although Onsager used the results in an article [29], he never published his dissertation.

Onsager spent 39 years of his career at Yale, where he was Assistant Professor of Chemistry (1934–1940), Associate Professor (1940–1945), and J. Willard Gibbs Professor of Theoretical Chemistry (1945–1972), a chair named after the father of statistical mechanics, the field of which Onsager was the greatest 20th-century authority. Until his retirement in 1972, he carried out research on chemical solutions, thermodynamics, and statistical mechanics with a distinguished group of theoreticians and experimentalists, of whom the most prominent include Raymond M. Fuoss, Julian Munson Sturtevant, Herbert Spencer Harned, Benton Brooks Owen, and John Gamble Kirkwood.

Because Onsager did not become a naturalized United States citizen until 1945, he could not participate in any war work during World War II. Instead, he spent the war years analyzing a problem in physics that many physicists thought to be insoluble—whether statistical mechanics could theoretically account for phase transitions of matter. By the use of obscure branches of mathematics such as quaternian and spinor algebra as well as the theory of elliptical functions, he demonstrated that the specific heat of a ferromagnetic system rises to infinity at the transition point. This solution of the so-called two-dimensional Ising model [30], first proposed by the Swedish physicist Gustaf Adolf Ising, is considered by many authorities to be Onsager's most spectacular achievement and one of the decade's most significant contributions to nuclear physics.

Onsager's interests were extremely wide, ranging from colloids and dielectrics to fractionation theory and superfluids. In his own words, "My field of interests is as unspecified as anybody's in the physical sciences" [31]. His contributions include an explanation of the Wien effect (the increase in the conductance of an electrolyte at very high potential gradients) (1934), a theory of thermal diffusion (1939), a theory of long-range orientational order in solutions of the tobacco mosaic virus (1948), the quantization of hydrodynamic circulation in liquid helium (1949), an explanation of the electrical conductivity of ice involving proton motion (1960–1974) [32], and flux quantization in superconductors (1961) [33]. He also studied the energy spectrum of turbulence (1945) [34], the statistical interpretation of the dissipation function (1953), a new interpretation of the Bose-Einstein condensation for interacting particles (1956) [35], electrical and magnetic behavior of metals in strong magnetic fields, the behavior of liquid crystals, order–disorder transitions, the statistics of unsteady flow in liquids, many-body problems, and the theory of metals. Negative absolute temperatures [36], which he first discussed in 1949, failed to attract much attention until the invention of optical pumping and lasers [37, 38]. Two international conferences were devoted to Onsager's discoveries—"Irreversible Thermodynamics and the Statistical Mechanics of Phase Transitions" (Brown University, 1962) and "Phenomena in the Neighborhood of Critical Points" (Washington, DC, 1965) [9].

Onsager held guest professorships at various universities. He spent a sabbatical year (1951–1952) as Fulbright exchange scholar with David Schoenberg (b. 1910), Director of the Royal Society Mond Laboratory of Cambridge University, a leading center for research in low temperature physics. His work there led to an explanation of the de Haas–Van Alphen effect (an effect occurring in many complex metals at low temperatures consisting of a periodic variation in the diamagnetic susceptibility of conduction electrons with changes in the component of the applied magnetic field at right angles to the principal axis of the crystal) [39]. He served as Visiting Professor of Physics at the University of California, San Diego (1961); Visiting Professor at Rockefeller University in New York (1967–1968); the Gauss Professor at the Universität Göttingen (1968); and Lorentz Professor at Leiden University (1970). In 1962, at the suggestion of German physicist and future (1967) Nobel Chemistry laureate Manfred Eigen (b. 1927), he joined Neuroscience Associates, a small interdisciplinary group in Cambridge, Massachusetts.

The Nobel Prize

Onsager received the Nobel Prize for Chemistry in 1968 "for the discovery of the reciprocal relations bearing his name, which are fundamental to the thermodynamics of irreversible processes." As we have seen, he had submitted the work on reciprocal relations in simultaneous irreversible processes under nonequilibrium conditions as a Norwegian abstract for a meeting of the Scandinavian Physical Society at Copenhagen in 1929. Stig Claesson called this contribution "a universal natural law, the scope and importance of which becomes clear only after being put in proper relation to complicated questions in border areas between physics and chemistry" [1a].

The final version was published in two parts in 1931 [21] but was ignored at the time and failed to attract attention until after World War II. When Onsager submitted it as a doctoral dissertation to the Norges Tekniske Høgskole, it was not accepted. Ironically, almost three decades later his *alma mater* awarded him an honorary doctorate (*Dr. Technicae*). According to Claesson,

> The elegant presentation meant that the size of the two papers was no more than 22 and 15 pages respectively. Judged from the number of pages this work is thus one of the smallest ever to be rewarded with a Nobel Prize [1a].

Although Alfred Nobel's will stipulated that the annual award of prizes referred to works "during the preceding year" [40], in Onsager's case a special rule of the Nobel Foundation was applied: "Work done in the past may be selected for the award only on the supposition that its significance has until recently not been fully appreciated" [1a].

Almost all common physical processes are irreversible and proceed in only one direction, for example, heat flows from a hot to a cold body, and a solute dissolves by mixing or diffusion. However, when a cold lump of sugar is dissolved in a cup of hot tea, these processes occur simultaneously, that is, heat is conducted to a cold body from a hot body, while sugar molecules diffuse throughout the tea.

Earlier attempts to deal with such processes met with little success when treated by classical thermodynamics, which had been developed during the 19th and early 20th century to study static states, that is, those at equilibrium, when no properties are changing. However, thermodynamics was not suitable for dealing with dynamic states that were not at equilibrium. The Second and Third Laws of Thermodynamics involved entropy, the measure of disorder in a system that provides a connection between thermodynamics and statistics. The American founder of thermodynamics, J. Willard Gibbs (1839–1903), who was Professor of Mathematical Physics at Yale and whose name was given to the professorial chair occupied by Onsager, applied statistical methods to the study of the random motion of molecules, thus making many of the crucial contributions to what became known as statistical thermodynamics [41].

Onsager applied these methods to study systems in which two or more irreversible processes, e. g., flows of heat or electricity, diffusion of matter, or chemical reactions of substances, are occurring simultaneously, and he developed the mathematics relating to the changing properties. He took the results that previous pioneers of thermodynamics had obtained for specific processes, and he generalized them in terms of symmetrical equations relating forces and flows within nonequilibrium systems.

To the three laws of classical thermodynamics, Onsager added two new principles—the principle of microscopic reversibility and the principle of least dissipation of energy [11]. He demonstrated that the reciprocal relations [21, 42] are mathematically equivalent to a more general principle of least dissipation, which stated that the rate of increase in entropy in coupled irreversible processes is at a minimum. A cogent summary of his prize-winning work without the inclusion of technical details was provided by Claesson:

> Onsager's great contribution was that he could prove that if the equations governing the flows are written in an appropriate form, then there exist certain simple connections between the coefficients in these equations. These—the reciprocal relations—make possible a complete theoretical description of irreversible processes.
>
> The proof of the reciprocal relations was brilliant. Onsager started from a statistical mechanical calculation of the fluctuations in a system, which could be directly based on the simple laws of motion which are symmetrical with regard to time. Furthermore he made the independent assumption that the return of a fluctuation to equilibrium in the mean occurs according to the transport equations mentioned earlier. By means of this combination of macroscopic and microscopic concepts in conjunction with an extremely skilful mathematical analysis he obtained those relationships which are now called Onsager's Reciprocal Relations [1a].

Onsager, as Claesson and others have proclaimed, was "far ahead of his time" [1a]. Irreversible thermodynamics, the core of his reciprocal relations—now sometimes called the Fourth Law of Thermodynamics—became fashionable only during the 1960s and is now universally recognized to be of immense value in chemistry, physics, biology, and technology.

Personality and *Modus Operandi*

As mentioned earlier, Onsager's interests were so varied that his graduate students, who were never numerous and were usually physicists rather than chemists, often came from entirely different scientific areas. Consequently, he never possessed a "research group," in the usual sense of the term, and he never founded a "school." Instead, he preferred to work out problems by himself rather than reading the solutions of others in the scientific literature, and he counseled his students to do likewise. He enjoyed working alone; almost all of his publications were written either by himself or with one collaborator.

Onsager read widely and translated several Norse epics into English. He was gifted with a phenomenal erudition, which colleagues sometimes found exasperating. C. T. Lane, the low-temperature physicist, once told him, "Lars, you don't know everything," to which Onsager replied, "But I'm learning" [10]. Onsager enjoyed gardening, swimming, and carpentry on his New Hampshire farm.

Onsager published only about one journal article per year, but they were always significant and frequently well ahead of their time, just like their author. He announced some of his discoveries in discussion periods at scientific meetings, sometimes in a cryptic manner; some of these he never tried to publish.

Several comments by colleagues provide insights into Onsager's personality and *modus operandi*. Regarding his theoretical abilities, Yale inorganic chemist Andrew Patterson, Jr. wrote,

> Perhaps what he is best at is making subtle mathematical substitutions which will collapse a formidable problem into a solvable one. This is real insight. In a time before computers became popular, he was able to make mathematical approximations which allowed problems to be solved that couldn't have been solved in any other way [43].

But Onsager's theoretical work also had profound experimental consequences. In the words of Yale physical chemist Philip A. Lyons,

> For the experimentalist, Onsager is a pure delight. His analyses of phenomena invariably define the practical scope of an experimental study and, what is more important, the direction of the study. Never has one of his theoretical proposals led to anything but substantial and fruitful research….Lars Onsager's contributions are those of a scientist with a marvelous blend of intuition and sophistication, zest and discipline. Though they differ widely in content, his papers have one thing in common: they offer a fresh look, usually at a vista [11].

According to Kirkwood, "Onsager's work is characterized not only by mathematical virtuosity but also by profound physical insight and intuition. His accomplishments are impressive and of lasting value [2]."

Onsager's office was a "windowless cubbyhole in the medieval-looking Sterling Chemistry laboratory" [11]. Longuet-Higgins and Fisher describe Onsager's first meeting with Joseph Hubbard, a postdoctoral fellow in 1971,

> On first setting eyes on his new postdoc, Lars embraced him in Russian style and took him to his office to show him a reprint. There was chaos on every surface, including the floor. Suddenly Lars disappeared, and Hubbard found him underneath the desk, where he had located the reprint (which turned out to be a 400-page thesis) and a two-month-old paycheck. Observing Onsager's contortions, Hubbard thought to himself: "Here's a fellow who scratches his left ear by reaching round the back of his head with his right hand. I wonder how he ties his shoes!" [8].

Honors and Awards

Onsager was elected to the National Academy of Sciences in 1947. He was a fellow of the American Association for the Advancement of Science, the American Physical Society, and the New York Academy of Sciences and a member of the American Chemical Society, the American Academy of Arts and Sciences, the American Philosophical Society, the Society of Sigma Xi, the Connecticut Academy of Sciences, the Bunsen Society for Physical Chemistry, the Royal Science Society in Uppsala, the Royal Society (London), the Institute of Physics (London), the Norwegian Academy of Technical Sciences, the Norwegian Chemical Society, and the Royal Norwegian, Royal Swedish, Royal Danish, and Royal Netherlands Academies of Sciences.

Recognition of Onsager's contributions did not come until relatively late in his life. In addition to the Nobel Prize, he received numerous honors, including the American Academy of Arts and Sciences' Rumford Gold and Silver Medals (1953); the Royal Netherlands Academy of Sciences' Lorentz Medal (1958); the American Chemical Society's G. N. Lewis (California Section), Willard Gibbs (Chicago Section), and John G. Kirkwood (New Haven Section, first recipient) Medals (1962), T. W. Richards Medal (Northeastern Section) (1964), and Peter Debye Award in Physical Chemistry (1965); Yeshiva University's Belfer Award in Pure Science (1966); and the President's National Medal of Science (1968). He held ten honorary doctorates: Harvard University (1954); the Norges Tekniske Høgskole (1960); Brown University, the Rensselaer Polyechnic Institute, and the Rheinisch-Westfählische Technische Hochschule, Aachen (1962); the University of Chicago (1968); Ohio State University (1969); Cambridge University (1970); Oxford University (1971); and Gustavus Adolphus College (1975).

Post-retirement Activities

As Onsager neared the retirement age of 68, the Yale Chemistry Department assured him that he could retain his office in the Sterling Laboratory of Chemistry. He had several research grants, making him able to count on continued governmental research support, including a salary, beyond retirement. However, Yale had a rule that an emeritus professor could not serve as principal investigator on research grants. Although Onsager asked that the rule be waived, his request was rejected by the administration.

Too offended to enter into negotiations, Onsager gave the administration several months to reconsider. When it failed to change its position, he accepted an offer to become Distinguished University Professor at the University of Miami's Center for Theoretical Studies, which allowed him to transfer his grants and have a group of postdoctoral research fellows and a secretary. There he continued to pursue consequences of previous research interests. For example, his work on proton movement in ice was a natural sequel to his more than four decades of work on conductivity of electrolytes.

At Miami Onsager turned his attention to biophysics. He became an associate of the university's Neurosciences Research Program and regularly attended its meetings. He demonstrated his profound knowledge of organic chemistry and biology in a paper that he presented at a 1968 Conference on Physical Principles of Biological Membranes [44]**.** The Center for Theoretical Studies arranged a symposium for his 70th birthday, at which many of his former students and postdoctoral fellows presented papers. His own contribution [45] speculated upon the origins of life on earth.

For years Onsager had suffered from phlebitis, a vein inflammation, but he remained an active scientist until the day of his death. He was found dead at the age of 72 in his home in Coral Gables, Florida on May 5, 1975 [5].

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