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The Origins of Onsager's Key Role in the Development of Linear Irreversible Thermodynamics

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Onsager discovered a deep underlying symmetry in the thermodynamic description of irreversible processes for which he was awarded the 1968 Nobel Prize in chemistry. After a brief sketch of irreversible thermodynamics and its history, this paper describes Onsager's path to his discovery. It is primarily based on an interview with Onsager a few months before his death, with some details from Onsager's Nobel address. When asked to rank his work, he placed this work at the top.

KEY WORDS: Onsager; Onsager reciprocal relations; history of irreversible thermodynamics; Onsager's ranking of his work; symmetry.

1. INTRODUCTION

For perspective, some background on linear irreversible thermodynamics$^{(2-4)}$ is helpful.

The real power of classical thermodynamics stems from the fact that only a small number of state variables are required to determine the properties of a uniform equilibrium system. To deal with irreversible processes in systems not too far from equilibrium, one divides the system into small subsystems and assumes that each subsystem is in *local equilibrium*, i.e., it can be treated as an individual thermodynamic system characterized by the small number of equilibrium variables.

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With this hypothesis, the entropy created *internally* by an irreversible process can be calculated. This quantity, ΔS_{int}, is a "balance" term to the Clausius inequality. Thus

$$\Delta S = \int \left(\sum \delta q_i / T_i \right) + \Delta S_{\text{int}}$$

where ΔS is the entropy change of the system (or subsystem) which must be calculated along a reversible path, and δq_i is the heat absorbed at boundary portion i of the system divided by the temperature T_i at that boundary portion, summed (or integrated) over the entire boundary of the system. The Second Law requires that ΔS_{int} be positive or zero. The "entropy production" σ is defined as the time rate of ΔS_{int} per unit volume.

For continuous systems, there is a T associated with each subsystem, and $T\sigma$ is the "dissipation," the dissipation function, or the "uncompensated heat" of Clausius. If $T\sigma$ is calculated for various irreversible processes, it is always found to have the form

$$T\sigma = \sum J_i X_i > 0$$

where J_i are flows of matter, heat, etc., and X_i are generalized driving forces like $d\mu/dx$ or $d\ln T/dx$ for vector transport processes or $\Delta\mu$ for chemical reactions, where μ is the chemical potential.

The J_i and X_i are linearly related when the system is not too far from equilibrium. Fortunately, "not too far" includes very steep gradients of concentration, temperature, and electric potential. Unfortunately, for chemical reactions "not too far" is too close to equilibrium to be useful.

Thus

$$J_i = \sum L_{ij} X_j$$

where the L_{ij} are called phenomenological, Onsager, or generalized transport coefficients. They are functions of T, P, concentrations, etc., but are not functions of the gradients X_i. In general the cross coefficients $(i \neq j)$ are *not* zero, are not always small, and can be positive or negative.

If the J_i are defined as the time derivatives of the extensive quantities of thermodynamics,(5) then the X_i in $T\sigma$ are determined. If the dependences are now removed among the J_i (e.g., using the definitions of reference frame) and among the X_i (e.g., using the Gibbs–Duhem equation), then there is an extremely important result due to Onsager(6, 7): the coefficient matrix of the L_{ij} for independent J_i and X_i is symmetric, i.e.,

$$L_{ij} = L_{ji}$$

These symmetry relations are called the Onsager reciprocal relations (ORR), and are verified by both theory(2) and experiment.(4, 5, 8), [2]

Because the formalism of this linear theory is the same for many different kinds of irreversible processes, the L_{ij} should be a more general way to examine and interpret experimental data. For example, in electrolyte solutions, diffusion coefficients, transference numbers, and electrical conductance are special case quantities. However, the more general ionic Onsager coefficients (L_{ij}) seem to be more useful for interpretation.(9–11) In conjunction with tracer diffusion coefficients, the L_{ij} have been found to provide a more sensitive test of electrolyte model theories than does the conductance.(12)

In summary, linear irreversible thermodynamics is based on the hypothesis of local equilibrium and the three fundamental concepts:

1. The dissipation function (irreversible production of entropy).
2. The linear relations between thermodynamic driving forces and flows of matter, electricity, heat, and of chemical reaction rates (near equilibrium).
3. Onsager's contribution, the symmetry of the coefficients of these linear relations between independent forces and flows.

2. ONSAGER'S RECIPROCAL RELATIONS

All three fundamental concepts had their origins in the 19th century, and had been described in isolated special cases. Dissipation function (entropy production) calculations go back to Rayleigh(13) (1873) for viscosity, Bertrand(14) (1887) for heat conduction, Duhem(15) and Natanson(16) independently (1896) for heat conduction and viscosity combined, and for more elaborate cases by Jaumann(17) (1911) and Lohr(18) (1916). Linear laws go back to Fourier(19) (1811) for heat conduction, Ohm(20) (1827) for electrical conduction, and Fick(21) (1855) for diffusion. Ad hoc variants of the equality of cross coefficients in the linear laws go back to Stokes(22) (1851) for anisotropic heat conduction, Kelvin(23) (1854) for thermoelectric effects, Helmholtz(24) (1878) for the equality of cell and Hittorf transference numbers, Saxèn(25) (1892) for electrokinetic effects, and Wegscheider(26) (1901) for chemical reactions.

Most of the ideas to provide a more general macroscopic theory were thus available by 1900, but no one saw the important unifying idea. That was left to Onsager's deep insight. He saw that the key idea was a systematic symmetry, and his insight was presented in two extremely significant papers in 1931.(6, 7)

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[2] Ref. 8 is a reprinting of ref. 4 plus new material.

In these papers, Onsager showed that the ad hoc assumptions of Stokes, Kelvin, Helmholtz, and of detailed balance for the triangle chemical reaction case could be related to a dissipation function. The corresponding linear laws had a characteristic, general symmetry which gave the earlier results as special cases. Onsager gave a statistical mechanical theory to justify the symmetry relations, based on the regression of fluctuations and on microscopic reversibility. The variants with time-reversal parity were also considered. A long paper with Fuoss in 1932(27) elaborated these ideas for transport in electrolyte solutions in terms of a Debye–Hückel model, in which diffusion and viscosity were also considered.

After 1932, Onsager went on to other problems and did little more along these lines, except for some statistical mechanical theory (with Machlup in the 1950s)(28, 29) and some pieces of the dissipation function in electrolyte transport theory (with Kim in 1957 and 1977(30, 31)).

Little attention was paid to Onsager's work for about 10 years. Then Josef Meixner, who had already gotten some results involving entropy balance and entropy production for thermoelectric effects in a magnetic field,(32) discovered Onsager's ideas. He found that they could systematize a large number of irreversible processes.(33–37) He worked out the current formalism, including the thermogalvanomagnetic effects and thermal diffusion. Subsequently DeGroot(38) and Prigogine(39) completed theses on these matters. They and their students, as well as others, began to explore the application of Onsager's ideas to a wide variety of linear phenomena. By the early 1960s, the macroscopic theory became "classic,"(2–4, 40, 41) since most of the interesting cases had been worked out and the validity of the ORR had been verified by experimental data for a wide variety of irreversible phenomena.(4, 5, 8) In 1968, Onsager received the Nobel Prize for the deep insight, originality, and generality of his contributions concerning symmetry in irreversible processes.

Although much current interest in the thermodynamics of irreversible processes lies in the nonlinear region, the classic linear theory provides an elegant general framework for describing linear processes. These are widespread, and include heat and electrical conduction in solutions and solids, thermogalvanomagnetic effects, diffusion, and electrokinetics, among others. In particular, the linear theory provides a complete foundation for multicomponent diffusion(42–46) and a very nice framework for the analysis of the properties of electrolyte solutions.(9–11, 47, 48)

There have of course been strident criticisms of applying its original statistical mechanical basis to vector processes, and petty arguments over definitions of "forces" and "flows."(49) Such quarrels over these issues are irrelevant with respect to the macroscopic theory, which can take the macroscopic Onsager–Meixner assumptions as "axioms." These axioms are subject to experimental verification, just as the laws of equilibrium thermodynamics are subject to such a test. As noted above, the validity of the Onsager reciprocal relations for linear phenomena is very well founded in experiment.(4, 5, 8) These issues, including the existence of other, more general statistical mechanical derivations, are discussed elsewhere.(5, 9, 46)

All this is background to the point of this paper. How did Onsager get his idea for unifying the symmetries observed in so many diverse phenomena? The story which follows is based on the author's interview with Onsager at Lawrence Livermore National Laboratory, March 14, 1976 (seven months before he died), as well as on his Nobel address.

In started in 1923. As a student, Onsager had previously come across the Helmholtz relation between the cell and Hittorf transference numbers(24) as well as the Kelvin relations of thermoelectricity.(23) These did not follow in a reasonable or convincing way from equilibrium thermodynamics, but seemed to be true.

After reading the first paper of Debye and Hückel on the equilibrium theory of dilute electrolyte solutions,(50) Onsager (still an undergraduate) began his early work on the transport theory of ions in solution. The results, based on Brownian motion kinetics, were obtained before the Debye–Hückel conductance paper was published.(51) He found the relaxation effect did not make sense unless it was treated symmetrically for conductance and transference. This effect was treated incorrectly by Debye and Hückel.(51) The hydrodynamic part of Onsager's theory could be based on a least dissipation principle, which would also involve symmetry. His papers involving this symmetry concept(52, 53) were written in Trondheim in the winter of 1925–26, after his famous visit to Debye. They were submitted later, and finally published in 1926 and 1927. Some of these first ideas were discussed with his professors at Trondheim, O. O. Collenberg and B. Holtsmark.

Nernst(54) had considered a kinetic theory of the diffusion potential, valid for dilute solutions, from which the Helmholtz transference number relation automatically came out. Onsager felt that the underlying assumptions of hydrodynamics, the symmetry of electrical or any other forces, together with least dissipation should give an overall symmetry. This belief ultimately led to the Onsager–Fuoss paper in 1932 for all the transport properties in electrolyte solutions based on a Debye–Hückel model.

The gradual development of this insight, that least dissipation was a unifying basis for symmetry, probably took place during 1924 but not later than 1925. It became clear during this period that the symmetry aspects of all kinds of irreversible processes, not just in electrolyte solutions, were all one problem. For example, Kelvin's analysis of thermoelectricity had assumed least dissipation after a fashion.

Chemical kinetics attracted his attention in 1924, based on the triangle reaction in the mutarotation of sugars (the experimental work of his professor, C. N. Riiber(55, 56)). Consideration of microscopic reversibility and detailed balance in chemical reactions also implied a symmetry. There seemed to be a least dissipation idea here, too.

Onsager was in Zürich from 1926 to 1928, working with Debye and Hückel. In the fall of 1927, he woke up one morning with a hangover, and got the idea for the general Debye–Hückel theory of electrolyte transport mentioned above. It would have symmetry and include diffusion and viscosity. After improvements, it was published with Fuoss in 1932.(27)

The idea of symmetry, obtained for electrolytes with a Debye–Hückel model and observed for other cases, came before a general proof of reciprocal relations. Onsager believed them to be right, but did not know how to prove it from first principles. The ideas for a general proof were ripening between 1926 and 1928. Finally he got the idea about fluctuations. What was really essential was the symmetry of past and future, i.e., microscopic reversibility. That concept had appeared in the special case of triangle chemical reactions. He outlined some of these ideas to P. Scherrer, who mentioned G. N. Lewis' considerable attention to microscopic reversibility. Since Lewis was a very important figure by then, Onsager said to himself that this hypothesis should be a first priority item for me. It had the advantage of being general enough to imply reciprocal relations in all kinds of irreversible processes in addition to chemical reactions.

At the same time, the success of least dissipation in guaranteeing symmetry in the hydrodynamics part of the electrolyte problem, its use in Helmholtz's ideas on the two transference numbers, and Kelvin's use of it in the thermoelectric case all implied a more general application as well.

All these ideas took a couple of weeks to a couple of months to gel in late 1927 and early 1928. In early February 1928, Onsager took a boat to go to the United States for a job in the Chemistry Department at Johns Hopkins University. While waiting for the boat, he had a couple of days in the port of Boulogne-sur-Mer. Here it struck him that there needed to be some connection with thermodynamics for the vector transport processes, just like the concentrations were for chemical kinetics. These in today's terms were the "thermodynamic forces," such as derivatives of chemical potential, etc. Of course there was a statistics of these. After a couple of months at Johns Hopkins, he began to formulate a first version of the theory of regression of fluctuations away from a state of maximum entropy. This resulted in a general principle of least dissipation and general symmetry relations. Once this was clear, "everything was clear." This was all worked out by the spring of 1928, but not in its final, simplest form.

However, it was the basis of the first announcement of the results in Norwegian in 1929.(57)

In the fall of 1928, Onsager started at Brown. In the summer of 1930, he attended a summer school on quantum mechanics at Ann Arbor, organized by Goudsmit and Uhlenbeck, which was attended by many renowned physicists. He immediately presented his results to Ehrenfest and a few other interested people. Ehrenfest thought the presentation was poor, but the content interesting. He suggested the final title of the 1931 papers, some hints on how to present them, that chemical reactions be given a prominent place, and that least dissipation should not be left out. He also gave Onsager a better idea about how to approach the statistical problem. Soon it occured to Onsager to formulate the statistical argument in terms of averages. The first of the papers,(6) with motivation, macroscopic descriptions, and a special case of the statistical argument, was prepared and submitted (received December 8, 1930; published February 15, 1931). The general statistical argument was presented in the second paper(7) (received November 9, 1931; published December 15, 1931). Notice how fast papers could be published 60 years ago.

Onsager also talked briefly about the work with Wigner and von Neumann at a Physical Society meeting in 1930, and to Kraus, Fuoss, Tamarkin, and Norbert Wiener at various times at Brown during 1930 and 1931. After the publication of the 1931 papers, there were a few inquiries, but not much interest.

As noted above, the idea for a general Debye–Hückel theory of electrolyte transport processes came to Onsager in 1927. In the fall of 1930, Fuoss came to Brown. He got interested in this problem, and the two of them started working on it in the winter of 1930. They jointly worked on the mathematics and the writing, but Onsager said the key simplifying ideas were his. This 90-page paper was published in 1932.(27) However, this was not Fuoss' Ph. D. dissertation. That was supervised by Kraus, was carried on in parallel, was on conductance in solutions of varying dielectric constant, and was finished in 2 years.

The subsequent history of irreversible thermodynamics was sketched above. Onsager simply put his technique in his "toolbox" to be withdrawn when convenient, such as for the work in thermal diffusion in 1939 with Furry and Jones(58) and the paper on diffusion in the Annals of the New York Academy in 1945.(59) He only glanced at the subsequent work of Meixner, Prigogine, DeGroot, and others, and did not really follow the working out of the details of what he had started. When asked, he said the macroscopic results were probably useful, when worked out in detail.

When I asked in this interview what his personal assessment was of his irreversible thermodynamics work, he said "Well, at the time that I did it, I told my mother that I think this is going to be the best thing I'll ever do." When asked did he still believe that, he said "Yes, although there might have been some other things that came closer than I hoped." When asked to state or write down what else was important and close, and to rank his work, he said, "Why would a guy want to do that." But on being pressed, and after a characteristic long silence, he mentioned the Ising model work.(60, 61) After another, still longer silence, he mentioned the quantization of vortices in liquid helium.(62) After a shorter silence, he mentioned the de Haas–van Alphen effect.(63) Then he mentioned the theory of dielectric constants of condensed phases(64) and said finally "of course the general work on electrolytes(65),[3] wasn't exactly peanuts; and a very tricky problem, the Wien effect in weak electrolytes,(66) turned out to be very useful."

Thus in this 1976 interview near the end of his life, Onsager felt that his work on these fundamental symmetry relations (the Onsager reciprocal relations) was his most important. As everyone knows, it got him the Nobel Prize. He ranked it ahead of all his other contributions, which followed the rank order: Ising model, quantization of vortices in liquid helium, de Haas–van Alphen effect, dielectric constants, and electrolyte theory. Some of these would also have deserved a Nobel Prize. There is no doubt that Onsager was one of the greatest scientific minds of this century.

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[3] See the extensive Longuet-Higgins and Fisher Bibliography(65) for the long list of electrolyte papers (27) from 1926 to 1977 by Onsager and his collaborators: R. M. Fuoss, N. N. T. Samaras, S. K. Kim, C. T. Liu, J. F. Skinner, S. W. Provencher, M. S. Chen, and J. B. Hubbard.

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