SPECIAL VOLUME IN MEMORY OF ILYA PRIGOGINE

ADVANCES IN CHEMICAL PHYSICS
VOLUME 135

Edited by

STUART A. RICE

Department of Chemistry
and
The James Franck Institute
The University of Chicago
Chicago, Illinois

AN INTERSCIENCE PUBLICATION
JOHN WILEY & SONS, INC.

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Wiley Bicentennial Logo: Richard J. Pacifico

Library of Congress Catalog Number: 58-9935

ISBN 978-0-471-68233-2

Printed in the United States of America

10 9 8 7 6 5 4 3 2 1

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INTRODUCTION

Few of us can any longer keep up with the flood of scientific literature, even in specialized subfields. Any attempt to do more and be broadly educated with respect to a large domain of science has the appearance of tilting at windmills. Yet the synthesis of ideas drawn from different subjects into new, powerful, general concepts is as valuable as ever, and the desire to remain educated persists in all scientists. This series, *Advances in Chemical Physics*, is devoted to helping the reader obtain general information about a wide variety of topics in chemical physics, a field that we interpret very broadly. Our intent is to have experts present comprehensive analyses of subjects of interest and to encourage the expression of individual points of view. We hope that this approach to the presentation of an overview of a subject will both stimulate new research and serve as a personalized learning text for beginners in a field.

STUART A. RICE

ACKNOWLEDGMENTS

The Symposium *In Memoriam Ilya Prigogine* on“Time, Irreversibility and Self-Organization” was held by the International Solvay Institutes for Physics and Chemistry at the University of Brussels on December 2–3, 2004.

This Symposium was made possible by the sponsorships of the European Commission, of the Fonds National de la Recherche Scientifique (FNRS), of the Nationale Fonds voor Wetenschappelijk Onderzoek (NFWO), of the Belgian National Lottery, of the “Communauté Franaise de Belgique,” and of the “Hotel Métropole,” which are gratefully acknowledged.

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CONTENTS

ILYA PRIGOGINE: HIS LIFE, HIS WORK*

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CONTENTS

*This chapter was originally published in the French language as "*Ilya Prigogine, sa vie, son oeuvre*, Mémoire in-8°, Classe des Sciences, Académie Royale de Belgique, 2006."

[†] Deceased, June 1, 2006

Special Volume in Memory of Ilya Prigogine: Advances in Chemical Physics, Volume 135,
edited by Stuart A. Rice

I. ILYA PRIGOGINE’S LIFE AND WORK

A. Introduction

In the history of science, there are few examples of such a flashing and immense ascent as that of Ilya Prigogine (Fig. 1). The little Russian Jewish immigrant arriving in Brussels at the age of 12 would end his life at age 87 with all the honors anyone—what is more, an intellectual—could dream of earning! Of course, the Nobel Prize in chemistry opened all the doors for him: He was able to use this opportunity for promoting a new vision of science. This extraordinary success is due, in the first place, to the importance of his works, but also to their novelty, to the introduction of the physicist to biology and the humanities, to his willingness to encourage dialogue, and to his leadership qualities in several international teams of researchers.

Figure 1. Photograph of Ilya Prigogine.

I had the privilege of being one of his oldest disciples and, subsequently, a researcher who remained close to Ilya Prigogine. In the present chapter, I wish first to relate the main events of his life (Section I.B). This presentation is based on my own actual experience and on my perception of Prigogine's personality. One will find another source (sometimes a little different in the interpretations!) in his autobiography, available on the internet:

www.nobel.se/chemistry/laureates/1977/prigogine-autobio.html

The second part of this chapter (Sections I.C–I.F) is devoted to an analysis of his work. Let me say immediately that, within the reasonable limits of this chapter, I could not cover the totality of his very diverse oeuvre. I thus made a selection, somewhat arbitrary, of course, but I tried to discuss its most characteristic aspects. I thus gave up the analysis of his works in quantum statistical mechanics (which are essentially a transposition and generalization of the central ideas of the classical theory), his work on vehicle traffic, his contributions to European science policy, and a few isolated papers. I do not present his works in chronological order, because of the strong entanglement in time of Prigogine's activities. Most often he was working on several problems at the same time. During some periods, one of the subjects would dominate, then another came to the surface, after which his interest would focus again on a problem that was shelved. I shall therefore divide my exposition into four groups of subjects, respectively, macroscopic physics and chemistry (Section I.C), microscopic physics (Section I.D), cosmology (Section I.E) and the philosophical aspects (Section I.F). The last section is an appendix, where I develop some more technical comments intended for the readers having a certain mathematical background. Finally, in Section II, I establish a systematic list of his numerous publications, classified according to their subjects.

B. Short Biography

Ilya Romanovich Prigogine was born on January 25, 1917 in Moscow. His father was a chemical engineer who owned a little soap factory, whose success was modest. His mother, Julia Wichman, had studied the piano at the conservatory. Young Ilya would inherit her love of music. It is said in his autobiography that he was able to read a music score before knowing how to read a book!

Ilya had an older brother, Alexander, who would become a chemist and make a career in the mining industry in the former Belgian Congo. He had a hobby that he developed very seriously: ornithology. He even discovered a novel species of bird, to which his name was attributed. After his return to Belgium, he was elected member of the ‘‘Académie Royale des Sciences d'Outremer'' (the Academy of African sciences).

The reader surely noted that Ilya Prigogine was born just a few months before the Russian revolution. Of course, the dramatic events of the time influenced unfavorably the business of his father. The family decided to leave Russia in 1921, spending a year in Lithuania, then going to Berlin, where young Ilya received a very good German education, which made him familiar with the classics of that nation. Ilya's father did not succeed in starting a new business in the disastrous economic circumstances of Germany of that time. Moreover, the threat of Nazism was visible on the horizon. The Prigogine family emigrated again and arrived in Belgium in 1929, where they settled for good. Young Ilya registered at the Athénée Royal d'Ixelles, an excellent Brussels high school. He had not yet made a final choice of a future career. On one hand, he was dreaming of becoming a professional pianist, but his teacher made him understand that he was not destined for that career.[1] Ilya also had a passion for history and archaeology (he realized later his dream by acquiring progressively an extraordinary collection of very rare pre-columbian objects). But finally, following his parents' and his brother's advice, in 1935 he started, in parallel, his studies of chemistry and of physics at the Université Libre de Bruxelles (ULB). He did not leave his Alma Mater until his death (even though he later committed some ‘‘infidelity’’ by accepting a part-time chair at the University of Texas at Austin).

During his studentship he continued to read the works of his favorite philosophers. The one who left a deep imprint on him and whom he would incessantly quote up to the end of his life was Henri Bergson.

The young student, aged 20, published in the *Cahiers du Libre Examen* (a local student journal) two papers: ‘‘*Essay on physical philosophy*’’ and ‘‘*The problem of determinism*,’’ followed by a third one, in collaboration with Hélène Bolle (who would become his first wife), ‘‘*The evolution*.’’ Remarkably, the roots of his future interests were already present in these works of his youth: determinism, the interpretation of quantum mechanics, biological evolution, and, above all, the concept of time.

Prigogine (who at that time signed his name as ‘‘Prigoshin,’’ a transliteration from Russian, doubtless inherited from his sojourn in Germany) then makes a fundamental decision, which will ‘‘put him in orbit’’: he chooses as director of his Master's thesis, and later his Ph.D. thesis, Professor Théophile De Donder.

The latter was a rather extraordinary person. Born in 1872, he started his career as an elementary school teacher, finally obtaining, as a self-taught man, the title of Doctor in physical and mathematical sciences in 1899. Very soon he became interested in relativity and started in 1916 a long correspondence with Albert Einstein. He also had contacts with many other great scientists of that

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[1] When the king of Belgium organized a reception at the Royal Palace on the occasion of the Nobel Prize, Prigogine asked the king to invite his old piano teacher: He performed some Chopin pieces. It was a very moving moment.

time: Henri Poincaré, Henri Lebesgue, Arthur Eddington, and more. As soon as he was appointed professor at the ULB, he started forming a remarkable generation of disciples, who would work at the ULB as well as abroad. Very soon he introduced the university to the most modern trends of physics of the time: relativity, quantum mechanics, statistical mechanics, thermodynamics. In the latter field, applied to chemistry, De Donder made an important advance by introducing the precise concept of affinity, which allowed him to calculate explicitly the entropy production in a chemical reaction. The door was half-opened toward nonequilibrium: Prigogine would open it widely.

Appointed as an assistant to De Donder in 1940, Prigogine had to give up this job when the University closed its doors in 1941, as a resistance to German occupation. The period of war was, of course, very hard. He was able to escape from persecution in the beginning, due to the organization of his fellow Russians, who provided him with documents certifying "officially" that he was a White Russian and was baptized. An unfortunate circumstance occurred in 1943: He was living with his companion Hélène Boll in an appartment previously occupied (without his knowledge) by a group of resistants, and thus he was in the focus of the Gestapo. The couple was arrested. During that same evening a dangerous expedition was organized by his friend Victor Mathot in order to recover in the apartment the manuscript of his future treatise on thermodynamics. Fortunately, the imprisonment was not very long: Due to numerous interventions, including one of Queen Elizabeth of Belgium in person, the couple was set free after a few weeks.

The war period was not one of scientific inactivity. The publication record of Ilya Prigogine contains 13 papers on thermodynamics published between 1940 and 1944 in the Bulletin of the Royal Academy of Belgium, in the Bulletin of the Chemical Society of Belgium, and in the Journal de Physique et le Radium (France). One learns from the acknowledgments of these papers that the young researcher was subsidized by the Solvay Institutes.

But Prigogine devoted these years mainly to the elaboration and the systematization of the ideas of his master De Donder. The result was his first "*magnus opus*" written in collaboration with Raymond Defay: *Treatise of Thermodynamics, in Conformity with the Methods of Gibbs and of De Donder*, whose first two volumes appeared in 1944 and 1946 (LS.3).

1944: ULB reopens its doors, and new opportunities are present. On september 13, De Donder writes a letter (whose original is preserved) to Prigogine. I am translating its second part[2]:

"3°) Chair of math. Chemistry: **applied** thermodynamics, **applied** stat. mech., **applied** wave mech., **applied energetics**.

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[2] The words in italics are underlined once, the words in boldface are underlined twice, the words in boldface capitals are underlined three times in the original handwriting.

In order to be appointed to this chair, it is *indispensable* to possess the title of **agrégé** *de l'Ens. sup.*[3] You must thus start immediately on the job. I believe that within *two months* you could, with the results already obtained (see **BOOK**, t. I, II, and III) and with the *new notes, in preparation, write a thesis of great originality and rich in important results.*

4°) Your memoir on the liquids would become your **ANNEX thesis**.

Your faithful, (s) Th. De Donder."

Ilya started working hard and obtained the title of *agrégé* in 1945. In 1947 he was appointed *Chargé de Cours* (≈ assistant professor), in 1950 *Professeur extraordinaire* (≈ associate professor), and in 1951 *Professeur ordinaire* (≈ full professor) at the Université Libre de Bruxelles, where he succeeded Jean Timmermans and was in charge of the course of Theoretical Physical Chemistry for the students in Chemistry.

In 1953 he was elected as Corresponding Member of the Royal Belgian Academy and becomes a full Member in 1960. He was at that time the youngest member of that Institution, where he developed an intense activity.

In 1959 Prigogine was appointed Director of the *International Institutes of Physics and Chemistry, founded by E. Solvay.* The main mission of these institutes, created in 1911 by the famous industrialist was the organization of the *Solvay Councils*, which gathered the greatest scientists of the time for discussions about the new major problems of science. Some of these Councils had a considerable historical importance. Thus, the Councils of 1911 and of 1927 were crucial moments in the birth of quantum mechanics; the latter witnessed the famous confrontation of Niels Bohr and Albert Einstein regarding the interpretation of the new mechanics. Prigogine gave new momentum to these institutes. While continuing the organization of the Solvay Councils, he widened the activities of the institutes by transforming them into high-level research institutes. Prigogine remained in this position until the end of his life.

Prigogine maintained strong international relationships with his foreign—in particular, American—scientists. Thus, he sojourned several times in the 1960s to the *University of Chicago* as a Visiting Professor, and he established long-living links with his colleagues. In 1967 he was appointed Professor at the *University of Texas at Austin.* His chair was later transformed into the "*Ilya Prigogine Center for Studies in Statistical Mechanics and Complex Systems*," to which he was appointed as the Director. Every year he would spend a significant fraction of his time in Austin. He would form there a team of scientists and teachers who would accompany him until the end of his life.

The supreme crowning of this intense activity was, in 1977, the award of the Nobel Prize in Chemistry to Ilya Prigogine, for his works in thermodynamics,

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[3] A post-doctoral title required in Belgium for an appointment as a University professor.

leading to the discovery of dissipative structures. As will be shown below, the existence of dissipative structures and their formation through bifurcations, made possible by the nonlinearity of the evolution laws, leads to the concept of creation of order by amplification of fluctuations. This concept of self-organization is central in biology, but also in sociology, economics, geography, and so on. The Swedish academicians were rightly inspired in awarding the Nobel Prize for this discovery, which plays an important role in chemistry and in physics, but also opens so many other doors. The dialogue, so much hoped for by Prigogine, between Science and Humanities clearly became possible in this framework. He effectively stimulated this dialogue through his interventions in the debates, colloquia, and other intercultural symposia which he organized.

From this moment on, Prigogine became a public person. He would make good use of this sudden popularity. He created a great tribune for the promotion of science in general, and of his own scientific and philosophical ideas in particular. Up to the end of his life, the signs of international recognition were accumulating. He was granted numerous international prizes rewarding his activity, both in science and in philosophy; he was awarded 54 honorary doctorates and was an elected member of numerous academies. He developed an intense activity as special counsellor at the European Union: Through his reports he contributed significantly toward a new orientation and momentum to the scientific policy of Europe. He also contributed to the opening toward the Eastern countries, particularly toward Russia. Institutes and high-level schools bearing his name have been created in Brussels, Austin, Moscow, Italy, and Argentina. He became a tireless traveler, transmitting his message around the world. One may also note during this period the numerous interviews given to the nonspecialized press and to television, both in Belgium and abroad.

I cannot finish this presentation without underlining his activity as a teacher. Up to his retirement in 1987, he influenced a large number of students at the ULB, mainly in chemistry. In the beginning he taught thermodynamics and the beginnings of quantum and statistical mechanics in third-year chemistry, and he delivered a specialized course on solutions in the fourth year. Later, his teaching acquired a much broader extension, covering “Theoretical Chemistry” over the whole undergraduate program. He was sharing his teaching duties with a strong team of associate professors (V. Mathot, F. Henin, C. George, G. Nicolis, R. Lefever, A. Goldbeter). In the physics curriculum, his course of theoretical physics was shared with P. Résibois and R. Balescu. The team was completed with a group of assistants who took care of the practical exercises. The style of *ex cathedra* teaching of Prigogine was quite singular. He loathed entering into details of calculations or into minutely detailed demonstrations (“you will see this at the exercises!”), thus transferring the burden to his assistants. On the other hand, he was great in providing an admirable overview of the subjects he treated. This would often lead him toward unexpected associations with music, philosophy, history, or neolithic art. This characteristic gave him a unique charisma, to which all his students responded.

But his teaching was not limited to his *ex cathedra* courses. His graduate students, researchers, and visitors had the privilege to participate in the discussions taking place in his office. Actually, Prigogine did not like working alone; he felt a strong need to share his ideas with his colleagues. (This explains why most of his scientific papers are published in collaboration with one or several authors.) He would then explain at length his latest ideas (again, without many mathematical details) and stimulate the reaction of his audience.

I would now like to evoke some more personal aspects of Prigogine's life. His first marriage with the poet Hélène Bolle (with whom he had a son, Yves) ended with a separation. After several years, during a visit in Warsaw in 1961, he met *Maryna Prokopowycz*. She was a chemical engineer and worked at the Warsaw University; it was love at the first sight.[4] Soon Ilya and Maryna married in Poland. But many months of strained waiting and numerous high-level interventions were necessary before the Polish authorities of that time authorized Maryna to join her husband. They became a very happy couple and had a son, Pascal. In the difficult moments, and also in the happy ones, Ilya could always count on the moral support—and especially on the love—his marvelous wife brought to him and which he brought to her.

All those who had the chance of knowing him remember in Ilya Prigogine a man of great generosity. He strongly supported and helped to form the careers of numerous researchers and teachers, not only in Brussels, but throughout the world: One finds disciples and admirers in Western and Eastern Europe, in Russia, in the United States of America, in Latin America, in Japan, in China, in India, and so on.

At the University his attitude was rather formal; for instance, he did not like to call his co-workers (even the old ones) by their first name (whereas he did this, necessarily, in America!). But privately, he would let down the barriers: I witnessed roars of laughter when he was feeling well. I also witnessed periods of anxiety, related to the lack of acceptance of his ideas (he mentions this also in his autobiography). But they were quickly overcome by his unfailing enthusiasm. He would sometimes tell me: "*Ah, Balescu, now we solved all the problems!*" And two years later: "*Ah, Balescu, now we* **really** *solved all the problems!*," and two years later, ...

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[4] In the following months we, the young co-workers in Brussels, started to consider his repeated travels to Warsaw rather "unusual," as were also the repeated invitations to Brussels of a Polish professor who delivered several seminars on the "powder electrodes," a subject of no interest to anyone here. His last seminar finished with an apotheose: He told us about the "powder electrodes without powder!" It was only later that we understood that this professor was Maryna's boss, and moreover that he would not let her leave Warsaw.

The last 10 years of his life were very painful. He was progressively sapped by an illness that made him suffer and that handicapped him physically. But up to the last moment he kept his mental readiness and continued following the work of his co-workers and suggesting new ideas.

He died in Brussels on May 28, 2003.

C. Macroscopic Chemistry and Physics

In Section I.A I mentioned the first book by Prigogine and Defay: *Traité de thermodynamique conformément aux méthodes de Gibbs et de De Donder* (LS.3).[5] At that time it was a quite original presentation of equilibrium thermodynamics, addressed mainly to chemists. The most striking feature is the absence of any reference to heat engines (Carnot cycles, etc.), which forms the starting point of all classical textbooks. The authors start from a concise, but simple and well-illustrated, exposition of the two principles. After that, there appears the basic notion of thermodynamic potential, from which the whole "treasure" of equilibrium thermodynamics follows logically. It is interesting to note (in order to understand the forthcoming subject) that practically the whole second half of the book is devoted to the properties of mixtures and solutions.

This treatise served as a basis for many teachers and was translated into numerous languages. Toward the end of his life, Prigogine felt the necessity of updating this treatise. He put his former co-worker Dilip Kondepudi in charge of this work, which he closely supervised. The result is a treaty: *Modern Thermodynamics, from Heat Engines to Dissipative Structures* (LS.15) (1998), which incorporates, in an attractive pedagogical form, all the progress achieved during the 54 years that separate it from the first version.

During the period 1945–1960, Prigogine worked on an intensive research program on **Mixtures and Solutions**. It can be framed into what can be called "Classical physical chemistry." It is clearly inspired by the professor he succeeded at the ULB and to whom many references are made: Jean Timmermans, a remarkable experimental physico-chemist. The results of these research efforts were published in a monograph written by Ilya Prigogine, Victor Mathot, and André Bellemans: *The Molecular Theory of Solutions* (LS.7), published in 1957; today this is still considered to be an important reference.

After this publication, Prigogine suddenly quit this research line (this was, however, continued by a group of his co-workers). One may wonder *a posteriori* what motivated the choice of this field of research, so singular in Prigogine's work. (Note, however, that his Master's thesis (1939) was already devoted to solutions of strong electrolytes, and, as noted above, half of his treatise on thermodynamics treats the same subject.) The "problem of time" that would

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[5] These symbols refer to the list of publications given at the end of this paper.

become Prigogine's obsession later is notoriously absent here. Was he, maybe, trying to get closer to the experimentalists? Whatever the answer, in a recent interview Prigogine declared about this period that "he did not regret anything!"

During the period 1945–1954 Prigogine continues to develop the project closest to his heart: **nonequilibrium thermodynamics**. His "thèse d'agrégation" (mentioned in Section I.B), *Etude Thermodynamique des Phénomènes Irréversibles* (1945) (LS.4), was the first book devoted exclusively to this subject. Whereas De Donder's works were devoted solely to the chemical reactions, Prigogine extended the formalism to all irreversible macroscopic processes, including transport phenomena in hydrodynamics and electromagnetism (diffusion, viscosity, thermal conduction, electrical conduction, and cross-effects, such as thermodiffusion). He derived the general expression (today, a classic!) of the entropy production, appearing as a bilinear form:

$$P = \sum_i J_i X_i \equiv \mathbf{J} \cdot \mathbf{X} \tag{1}$$

where J_i denotes the set of dissipative *fluxes* (e.g., matter fluxes, heat flux, chemical reaction rates), and X_i denotes the corresponding thermodynamic *forces* (e.g., density or concentration gradients, temperature gradient, chemical affinities). [The set of fluxes (J_i) and of forces (X_i) can be grouped into "*vectors*" **J** and **X**, which lead to more compact formulae.] This formula is very general, being valid in the whole domain where the macroscopic equations of evolution are valid. The fluxes and the forces are interrelated by phenomenological transport equations. During this first period, Prigogine limited himself to the simplest case, where these equations are *linear*. He then derived his celebrated theorem of *minimum entropy production in the nonequilibrium stationary states*, which is valid precisely in this linear domain.[6] The latter is even applied in biology, in a paper by Prigogine and Wiame (THL.8), where the authors conjecture that the living systems (necessarily open systems, exchanging matter and energy—and entropy!—with the external world) evolve toward a state of minimum entropy production. This evolution goes together with a global decrease of entropy, thus toward a complexification, creating structures. This paper contains the first attempt of application of nonequilibrium thermodynamics to biology.

The "thèse d'agrégation" ends with a very brief chapter "*Time and entropy*," which contains the root of Prigogine's future preoccupations. He defines a "*thermodynamic time*" related to the entropy production. It is interesting to point out one of the last conclusions of this chapter: "*Originating from the second principle, the thermodynamic time necessarily appears as a statistical concept. It loses its meaning at the scale of elementary processes*." This

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[6] One may note that Prigogine inherited from his master, De Donder, his love of variational principles!

conclusion will be vigorously repudiated 25 years later (when Prigogine will insist on the universality of the irreversible time, on all scales; see Appendix).

In the following years, Prigogine developed various additional aspects of the new thermodynamics. He published in 1955 the little treatise "*Introduction to Thermodynamics of Irreversible Processes*" (LS.6), which was very successful and was translated into many languages.

The year 1954 is a landmark in Prigogine's research in thermodynamics: for the first time he ventures to break down the "barrier of linearity." As in all sciences, the simplest problems occur when one studies the phenomena that happen in the neighborhood of a known reference state. The tools necessary for this study have been handed down to us by the mathematicians of the nineteenth century: linear analysis, a complete, simple and elegant formalism, offering the solution of all problems in this realm. Unfortunately, when applied to physical problems, one must take into account that its validity range is very limited.

Beyond, one enters a "*terra incognita*," where all surprises are possible. Prigogine entered it resolutely, accompanied in the beginning by his old friend Paul Glansdorff. The latter, also a disciple and admirer of De Donder and a few years older than Ilya, was a man with a very warm character; he was also extremely refined and was erudite in French history of science. He developed first a career as an engineer, exploiting his mastery of thermodynamics (applied to the industry of refrigeration); he was at that time a professor at the Polytechnic Faculty in Mons, Belgium. Prigogine obtained his appointment at the ULB; from that time on, a very fruitful collaboration started between the two men. The problem that was open can be formulated as follows: *How can one describe and study the irreversible phenomena occurring in a system in which the relations between fluxes and forces are no longer linear, or even no longer exist (as univocal functions)?* Glansdorff and Prigogine tackled the problem using the variational methods, dearest to the heart of their former master De Donder.

In an important paper (TNC.1), they offered for the first time an extension of nonequilibrium thermodynamics to nonlinear transport laws. As could be expected, the situation was by no means as simple as in the linear domain. The authors were hoping to find a variational principle generalizing the principle of minimum entropy production. It soon became obvious that such a principle cannot exist in the nonlinear domain. They succeeded, however, to derive a "half-principle!" They decomposed the differential of the entropy production (1) as follows:

$$dP \equiv d_J P + d_X P = \mathbf{X} \cdot d\mathbf{J} + \mathbf{J} \cdot d\mathbf{X}$$

and then proved that

$$d_X P \leq 0 \tag{2}$$

This principle is very general, relating neither to the linearity nor to the symmetry of the transport laws. On the other hand, it is difficult to attribute a physical meaning to $d_X P$. The authors later attempted to derive a "local potential" from this property, and they applied this concept to the study of the chemical and hydrodynamical stability (e.g., the Bénard convection). The results of this approach were published in Glansdorff and Prigogine's book: *Thermodynamic Theory of Structure, Stability and Fluctuations* (LS.10, 10a), published in 1971.

In two papers of 1954 and 1956 (TNC.3, 5) Prigogine and Balescu showed that the property (2) opened the possibility of existence, far from equilibrium, of *oscillating chemical reactions*. It was a statement that was highly "politically incorrect" at that time. In a subsequent paper (1959), Thor Bak concluded: "*It is pointed out that none of the chemical reactions alleged to show oscillatory behavior have been thoroughly investigated experimentally*!" But in fact, in 1958 in an obscure Siberian journal, there appeared a paper by Belousov where the author announces the discovery of a true chemical clock; this work was taken over in 1964 by Zhabotinsky, and the news became known in the West. It was a marvelously simple reaction: One put together in a test tube some appropriate reactants, and one witnessed a change of color of the liquid, turning from red to blue and back within a period of a few minutes, thus easily observable and without any sophisticated equipment. The theoretical predictions of Prigogine and his co-workers were thus admirably confirmed by experiment! Later, many oscillating chemical and biochemical reactions were discovered and studied.

The year 1967 appears as a crucial year: In an important paper by Prigogine and Nicolis, "*On symmetry-breaking instabilities in dissipative systems*" (TNC.16), there appears for the first time the term "dissipative structures." The filiation of this concept with the "half-principle" of Glansdorff and Prigogine can be clearly perceived in the works of that period (particularly in the paper TNC.17). However, the new approach required a radical change of the theoretical methods.

In their subsequent works, the authors treated directly the nonlinear equations of evolution (e.g., the equations of chemical kinetics). Even though these equations cannot be solved explicitly, some powerful mathematical methods can be used to determine the *nature* of their solutions (rather than their analytical form). In these equations, one can generally identify a certain parameter κ, which measures the strength of the external constraints that prevent the system from reaching thermodynamic equilibrium. The system then tends to a nonequilibrium stationary state. Near equilibrium, the latter state is unique and close to the former; its characteristics, plotted against κ, lie on a continuous curve (the thermodynamic branch). It may happen, however, that on increasing κ, one reaches a critical **bifurcation** value κ_c, beyond which the appearance of the curve suddenly changes. For $\kappa > \kappa_c$ a branching of trajectories occurs, and mutiple stationary states appear, some of them stable, others unstable. The system then has the possibility of *choice* of proceeding along one or the other of these curves. Another possibility (Hopf bifurcation) is the appearance, beyond κ_c, of a *limit cycle*. One then witnesses an oscillating behavior (like the one produced in the Belousov–Zhabotinsky reaction). In all these cases a **temporal symmetry breaking** occurs: The character of the evolution is radically different from the one present in the neighborhood of equilibrium. Last but not least, when inequalities of concentration exist in various parts of space, diffusion enters the game. In that case, the bifurcation may lead to stationary states that are spatially structured (for instance, spatially periodic): Here the **symmetry breaking is spatial** as well as temporal. One finds in all these cases the appearance of states whose properties are totally different from those of equilibrium, and which can only live at finite distance from equilibrium. Prigogine and Nicolis call these states **dissipative structures**. The latter must necessarily ‘‘feed’’ on fluxes of matter and/or energy (thus, on external constraints) that permanently maintain the system far from equilibrium. They can therefore only exist in open systems. All these ‘‘bizarre’’ phenomena (and many others) are consequences of the *nonlinearity of the evolution laws*, and possibly of the competition between nonlinearity and (linear) spatial diffusion. When one goes even farther from equilibrium, new secondary, tertiary,. . ., and so on, bifurcations may occur, leading to new structures and possibly to a transition to chaos.

In parallel with the studies described above, which concern perfectly deterministic equations of evolution, it appeared necessary to complete the theory by studying the spontaneous **fluctuations**. Near equilibrium, any deviation is rapidly damped; but near a bifurcation point, a fluctuation may may lead the system ‘‘across the barrier.’’ The fluctuation is then stabilized, or even amplified: this is the origin of the phenomenon which Prigogine liked calling ‘‘*creation of order through fluctuations*.’’ More specifically, one witnesses in this way a step toward *self-organization.*

Once the door was opened to these new perspectives, the works multiplied rapidly. In 1968 an important paper by Prigogine and René Lefever was published: ‘‘*On symmetry-breaking instabilities in dissipative systems*’’ (TNC.19). Clearly, not any nolinear mechanism can produce the phenomena described above. In the case of chemical reactions, it can be shown that an *autocatalytic step* must be present in the reaction scheme in order to produce the necessary instability. Prigogine and Lefever invented a very simple model of reactions which contains all the necessary ingerdients for a detailed study of the bifurcations. This model, later called the ‘‘*Brusselator*,’’ provided the basis of many subsequent studies.

In 1969 a paper by I. Prigogine, R. Lefever, A. Goldbveter, and M. Hershkowitz-Kaufman was published: ‘‘*Symmetry-breaking instabilities in* *biological systems*" (TNC.21) started a new direction of research that later proved very fruitful. In living systems one finds many oscillating chemical reactions that determine the fundamental rhythms of the organisms. The spatial symmetry-breakings might explain the formation of biological structures. In fact, the first forerunning work on dissipative structures is due to the great British mathematician Alan Turing, who, in 1952, established a model of morphogenesis involving such a symmetry breaking. The same spatial symmetry-breaking may (perhaps?) provide a possible explanation to the origin of life.

The subsequent 10 years were filled with an intense activity developing the new ideas and seeking new applications in progressively wider fields, including problems of economy, sociology, and geography. (Let us quote, for example, the very original works of Prigogine, Peter Allen, Françoise Boon, and Michèle Sanglier in the problem of urban development). These results were collected and synthesized in a remarkable book by Nicolis and Prigogine: *Self-Organization in Non-equilibrium Systems* (1977, LS.12); this matter was completed and updated in 1989 in *Exploring Complexity* (L.S.14) by the same authors. All these works were rapidly recognized and further developed by the international community of physico-chemists.

D. Microscopic Physics

In parallel with his work in thermodynamics, as soon as 1950, Prigogine took on the problem of the *microscopic foundation of irreversible phenomena*. The latter path, full of pitfalls, involving long fruitful periods, interrupted by periods of stagnation, or even of reversals, would preoccupy him for the next 50 years, until the end of his life (200 papers and several books). A communication to the Belgian Society of Logic and Philosophy of Science, delivered in 1951 under the title "*Probabilities and Irreversibility*" (GEN.5), is particularly illuminating. After an exposition of the paradox of irreversibility (irreversibility of macroscopic phenomena, but reversibility of the microscopic dynamical laws), Prigogine criticized the approaches of Ehrenfest and of Kirkwood, based on an operation of "smoothing" (*coarse graining*) in phase space. He concluded by establishing a program that would be his own for the remainder of his life:

> "(One asks) three questions:
>
> (1) What mechanism explains the independence of the final distribution with respect to the initial distribution . . .?
> (2) What is the relaxation time of the distribution, i.e., the time necessary for the establishment of the latter independence?
> (3) How do the externally imposed constraints (temperature gradient, bulk velocity,. . .) modify the asymptotic distribution?

We may only raise these questions. The development of the main mathematical tools allowing their study (theories of Markoff chains and of linear operators in Hilbert space) may, however, allow us to hope for progress in the coming years."

A posteriori, one may insert "50" before "years"!

The first attempts (G. Klein and I. Prigogine, 1953, MSN.5,6,7) were very timid and not very conclusive. They were devoted to a chain of harmonic oscillators. In spite of a tendency to homogenization of the phases, there was no intrinsic irreversibility here, because an essential ingredient is lacking in this model: the interaction among normal modes. The latter were introduced as a small perturbation in the fourth paper of the series (MSN.8).

In 1955 a fundamental paper by *Léon Van Hove* (a great physicist, Prigogine's friend, former student at the ULB, and, at that time, professor at the University of Utrecht, Netherlands) was published. The paper was devoted to weakly coupled many-body quantum systems. The author realized the first sharing of the recently developed mathematical methods of quantum field theory (renormalization) with those of statistical mechanics. This sole aspect (beyond the importance of the results obtained) would suffice to establish the importance of this work: It was going to "refresh" statistical mechanics, by introducing a new "toolbox," which would become indispensable. In his work, Van Hove underlined the fact that even very weak interactions (measured by a parameter $\lambda \ll 1$) lead to contributions that grow limitlessly for long times ("secular terms"). It is thus necessary to perform a perturbation calculation *to all orders in* λ, select in each order the most divergent terms, and resume the resulting partial series in order to obtain a globally finite result.[7] Van Hove succeeded [by retaining the contributions of order $(\lambda^2 t)^n$] in deriving in this way Pauli's (irreversible) equation of evolution, thus avoiding the *ad hoc* assumptions introduced by earlier investigators.

This work (actually very difficult to read, and using a very heavy formalism) had the effect of a bomb in Brussels. Prigogine associated himself with Robert Brout (who was at that time a postdoc in Brussels) in order to understand, deepen, and develop Van Hove's ideas. The first result of this collaboration was a basic paper (1956, MSN.12) on the general theory of weakly coupled classical many-body systems.[8] Although still influenced by Van Hove's paper, this work by Brout and Prigogine is a generalization of the latter, as well as a simpler and more transparent presentation.

[7] As a trivial example, think of the exponential function $\exp(-t)$ for positive times: Its series expansion contains all positive powers of time, $t, t^2, t^3, \ldots$, which grow indefinitely for a long time; the sum of their series, however, is finite and decreases to zero.

[8] This paper was published as Part VII (and last!) of the series "Statistical Mechanics..." initiated by Klein and Prigogine. It represents, however, a radical change with respect to the preceding papers of the series!

Without discussing details (some of which are discussed in the Appendix), I should like to underline the fact that one can identify in this paper the starting point of an idea that Prigogine would pursue in all subsequent years. The classical problem of mechanics is based on the representation of the instantaneous state of the system by specifying the coordinates and the momenta of all its particles. This state is thus represented by a *point* in the many-dimensional "*phase space*." By specifying an initial condition—that is, the position of the representative point at a given time, as well as the forces that operate in and on the system—Hamilton's equations of motion determine the position of the point at any other earlier or later time. In other words, these equations determine a unique curve in phase space: the *trajectory* of the system. But, in quantum mechanics (because of Heisenberg's uncertainty principle), and also in classical mechanics of "nonintegrable" systems, it is shown that such a specification is impossible (or, at least, illusory). On the other hand, the study of an *ensemble* of systems (obeying the same equation of motion, but with different initial conditions), described by a *probability density* in phase space, briefly called a *distribution function* ρ, is perfectly univocal and leads, through Liouville's equation of evolution, to a statistical description of classical systems.[9] This concept would become the basis of the young science of nonequilibrium statistical mechanics. The idea was far from being new: It was introduced in the beginning of the twentieth century by Gibbs. Prigogine, however, had already in 1950 considered the "*death of trajectories*" to be an intrinsic property of unstable dynamical systems which, alone, would lead to an understanding of irreversibility. It will become the main axis of his theory and, more generally, of his vision of the world. The development of these ideas is briefly sketched in the Appendix.

Robert Brout would soon go back to the United States, but already a group of young enthusiastic students and future Belgian scientists was growing around Ilya Prigogine; they would develop and amplify the new nonequilibrium statistical mechanics. Among the earliest, in order of arrival, let me list them: Radu Balescu, Françoise Henin, Pierre Résibois, Claude George. It is interesting to note that we were all chemists, converted to physics by the charisma of Prigogine. The period 1956–1970 was certainly one of the most fertile for the Brussels group.

The research was greatly facilitated by two important elements. The (formal, perturbative) solution of the Liouville equation is greatly simplified by a Fourier representation (see Appendix). The latter allows one to easily identify the various types of *statistical correlations* between the particles. The traditional dynamics thus becomes a *dynamics of correlations*. The latter is completed by

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[9] In the present paper I shall only discuss explicitly classical systems. The extension of these methods to quantum systems is possible, and it has been done by Prigogine and his co-workers.

the elaboration of a *diagram technique* (MSN.25) that establishes a correspondence between the numerous terms of the perturbation expansion and some well-adapted and efficient diagrams. It thus becomes possible to estimate visually, before any calculation, the order of magnitude of any term in the perturbation expansion of the distribution function. [The idea of a diagram technique was previously introduced in quite different fields: Mayer in the 1930s, for equilibrium statistical mechanics, and Feynman in the 1950s, for quantum field theory]. From here on, the machine could start moving. In the 1960s, Prigogine and/or his collaborators obtain new kinetic equations for systems as varied as: anharmonic solids (Henin), plasmas (Balescu), liquids (Nicolis, Misguich), quantum gases (Résibois), ferromagnets (Résibois), gravitational systems (Severne), relativistic systems (Balescu), scattering theory (Mayné), and so on. At the same time, the method provided a sound theoretical basis for macroscopic physics.

The vision of irreversibility that appeared in this first group of works, which formed the object of the first monograph on nonequilibrium statistical mechanics by Prigogine (1962, LS.9), was the following. The necessary conditions for an irreversible evolution were:

1. The large size of the system (i.e., a large number N of particles) enclosed in a large volume V. The calculations are greatly simplified in the thermodynamic limit: $N \to \infty$, $V \to \infty$, $N/V = n$: *finite*.
2. A (somewhat technical) condition on the resonances.
3. Finite range of the interactions and of the correlations.
4. The presence of a "small parameter."

Under these conditions, the distribution of the action variables (e.g., the momenta) [the *vacuum*, ρ_0] tends irreversibly toward the thermodynamic equilibrium after a sufficiently long time. Under the same conditions, the correlations are determined by the vacuum (technically, they become functionals of the vacuum distribution ρ_0) (see Appendix).

It should be stressed that in this first group of works (1956–1970) *there is nowhere any deviation from the Hamiltonian laws of dynamics*. This feature was taken as a preliminary postulate of our work. Irreversibility appeared as an **asymptotic** property of the evolution of certain classes of systems. The term "asymptotic" refers to the large size of the system, as well as to the long time scale of observation.

After 1969, the "Prigoginian" statistical mechanics started to change its aspect. I shall try to outline here the chronology and the significance of these changes. The more technical aspects will be discussed in the Appendix.

A first purpose consisted "only" of generalizing the domain of validity of the theory developed during the years 1956–1970. Prigogine's ambition was to show that the irreversible behavior found in the simple dynamical models was not limited by the approximations related to the presence of a small parameter (weak coupling, weak density,...). In all those cases, the existence of widely separated characteristic time scales leads to the representation of irreversibility as an *asymptotic* property, manifesting itself after a sufficiently long time, compared to the shortest characteristic time scale (e.g., the duration of a collision). In what way could one get rid of the restriction to small values of the characteristic parameter and generalize this behavior to systems with arbitrarily strong coupling?

A first answer to this difficult question was elaborated from 1966 on. A new methodology was introduced by C. George in 1967. It consisted of identifying a "piece" of of the distribution function that would evolve irreversibly to equilibrium, following a "**subdynamics**," *independently of its complementary part*. This identification is made operational by introducing a set of **projectors**, leading to a new, very elegant formulation of statistical mechanics.

By that time, a new member, of sizable stature, appeared in the Brussels group. Near the end of his life, *Léon Rosenfeld* left Copenhagen (where he had been for many years the right-hand man of Niels Bohr) and came back to his native Belgium. He became converted to Prigogine's ideas, which he actively supported and of which he became an enthusiastic promotor.[10] This collaboration became concrete in 1973 in a long review paper by I. Prigogine, C. George, F. Henin, and L. Rosenfeld (**PGHR**): "*A unified formulation of dynamics and thermodynamics*" (MSN.75).

The first motivation of Prigogine (which guided him actually since the beginning of his researches in this field) was to obtain a *general microscopic definition of entropy*, the universal indicator of irreversibility. This is a crucial question for establishing the molecular basis of thermodynamics. It was stated, and solved in the special case of dilute gases, by Ludwig Boltzmann in 1872. But this solution required a partial abandonment of the laws of mechanics, along with entering into the game of **probabilities**. The result was an avalanche of criticisms from the supporters of a purely deterministic evolution (i.e., the majority of the physicists of that time). In spite of this, "it worked!": Boltzmann's theory led later to a determination of the macroscopic transport coefficients of gases with a great precision, and it was fully supported by experiment.

The generalization of Boltzmann's solution turned out to be especially difficult. In their 1973 paper, PGHR performed a synthesis of the *projector* method of C. George and the idea of a **transformation** of ρ. The PGHR paper was considered for several years as the "bible" of Prigogine's group. The

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[10] A significant anecdote: During a summer school in Sitges, Spain, in 1972, an anonymous student replaced the lettering of the announcement of a lecture by Rosenfeld about Prigoginian statistical mechanics by substituting the anagram: "EL DEFENSOR"!

authors developed a theory leading to a “causal irreversible dynamics” and to a definition of entropy. When the Indian physicist *Bandyanath Misra* arrived in Brussels, the final touch was achieved for this construct. For this reason, in order to avoid repetitions, I prefer to discuss here only this final form of the theory.

The new formulation, which approached the problem from a totally different point of view, appears in a series of papers, from which we extract mainly: *B. Misra, I. Prigogine, and M. Courbage* (**MPC**) (1979, MSN.98) and *S. Goldstein, B. Misra, and M. Courbage* (**GMC**) (1981, *J. Stat. Phys.*). The set of all Prigogine’s works on macroscopic dissipative structures and on microscopic dynamical systems are the object of a review in the book “*From Being to Becoming*,” published in 1981.[11] The presentation is “semitechnical,” intended for readers having a basic mathematical and physical background, without, however, entering the details of the derivations. The book was conceived as a complement to *La Nouvelle Alliance*, and therefore it inscribes the scientific results into a more general philosophical framework.

Given the importance of the work of Misra, Prigogine, and co-workers, I am providing in the Appendix a detailed, but simplified, analysis. The main result of these works can be concisely formulated as a unique *theorem*:

> There exists a class of dynamical systems, whose distribution function ρ obeys the (deterministic) Liouville equation, for which one can prove that, as a result of a transformation Λ of ρ, their evolution toward the future is “similar” to an irreversible stochastic evolution towards equilibrium, obeying a probabilistic evolution law. The transformed distribution function leads to a simple definition of an entropy. The members of this class will be called **intrinsically stochastic dynamical systems**.

A few remarks will help us in understanding the importance of this result. One should first stress the fact that this proposition (when precisely formulated) has the status of a *theorem*, proven with the full rigor required by mathematics. Next, one should note that we have here an “existence theorem”: The class of systems to which it applies is defined in a univocal, but abstract, non-constructive way. In the paper GMC, the authors show that the necessary and sufficient condition of intrinsic stochasticity is that the systems belong to the class of “*Kolmogorov flows (K-flows)*.” Although this class is mathematically well-defined, it is extremely difficult to prove that a given *physical* system belong to this class. On simplifying to the extreme, it may be said that these are systems exhibiting a high degree of *dynamical instability.* For these systems, a very small variation of the initial condition may lead to enormous deviations after a finite time.

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[11] The Italian version, “*Dall’essere al divenire*,” of 1986, is considerably updated and includes in particular the MPC theory.

Note also that the formulation of the theorem contains the following restriction: "evolution toward the future." This implies that the evolution "toward the past" is described by a different law (splitting of the evolution Group into two irreversible Subgroups). Prigogine calls this feature a "*temporal symmetry-breaking*," related to the "*arrow of time*." (Unfortunately, he fails to state that this "symmetry-breaking" has in no way the same meaning as the one found in the macroscopic dissipative structures: Here there is no critical threshold, no bifurcation, no multiple stationary states, and so on.)

It is important to stress the fact that in the proof of the MPC theorem, *the laws of classical dynamics are never violated*. One could summarize the significance of the MPC theorem by saying that, *for a well-defined class of dynamical systems, the new formulation "lays bare the arrow of time" that is hidden in the illusorily deterministic formulation of these unstable systems*.

The intrinsically stochastic systems are defined in an abstract way: It could be said that they live in the world of Platonic ideals. In order to transform this *mathematical* theory into a *physical* theory, one should be able to prove that there exist material systems satisfying the instability criteria required by MPC. At present, however, such a demonstration (satisfying the same criteria of mathematical rigor) does not exist. Therefore, in order to illustrate the consequences of their theory, MPC made do with a dynamical system reduced to its simplest expression: the "*baker's transformation*."[12] In this purely mathematical model, all desired quantities can be exactly and explicitly calculated; this was done by Prigogine and his co-workers for many coming years. The baker's model thus became the paradigm of the intrinsically stochastic systems. It is discussed in detail in all the books published by Prigogine in the following years.

But the major physical problem remained open: Could one prove *rigorously* that the systems studied before 1979—that is, typically, systems of N interacting particles (with N very large)—are intrinsically stochastic systems? In order to go around the major difficulty, Prigogine will take as a starting point another property of dynamical systems: **integrability**. A dynamical system defined as the solution of a system of differential equations (such as the Hamilton equations of classical dynamics) is said to be *integrable* if the initial value problem of these equations admits a unique analytical solution, weekly sensitive to the initial condition. Such systems are mechanically stable. In order to

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[12] In this model the continuous evolution in time is replaced by successive transformations of the space on itself ("mappings"). The baker mapping consists of starting from an $L:L$ square and stretching it in one dimension while shrinking it in the other dimension, thus forming a rectangle $2L:L/2$; one then folds the latter on itself, in order to make again a square $L:L$. It is easily conceivable that after sveral transformations a finite region in the interior of the initial square is deformed, fragmented, and dispersed in the final square: This is the property called "*mixing*."