

Pierre Résibois: 1936–1979

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Main aspects of Résibois' scientific work.

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The physics community was deprived of one of its distinguished members by the premature death on 9 February 1979 of Pierre Résibois. He was born in Brussels in 1936 and received his PhD degree in 1960 from the University of Brussels. He was associated for six years with the Belgian National Science Foundation (F.N.R.S.). In 1966, he became an Associate Professor at the University of Brussels. He also taught at Leiden University and Harvard University. He was a member of the IUPAP Commission in Thermodynamics and Statistical Physics and was on the editorial board of the *Journal of Statistical Physics* and *Physica*. His main research interest was in nonequilibrium statistical mechanics.

The main characteristic of Résibois' scientific work is a deep and sure physical intuition. Relying on this intuition he developed elegant and powerful methods to study the irreversible behavior of N -body systems. For example, his factorization theorem^{[26],2} offered a useful tool for the derivation of "one-time" kinetic equations in quantum systems.^[39] Moreover, he was not only interested in deriving formal kinetic equations; indeed, he also applied these sophisticated techniques to concrete problems in order to obtain results or to make predictions which could be compared with experiments. For instance, he was among the first to make an explicit

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² Reference numbers in square brackets refer to the list of Résibois' publications given at the end of this paper.

evaluation in the critical region of the correlation functions which are determined in the neutron scattering experiments on magnetic systems.

Since the early days, nonequilibrium statistical mechanics has known many controversies. In these debates, Résibois was a tenacious fighter for the theories he held to be of value. One of us (I.P.) remembers that when as a graduate student Pierre attended a summer school in the Netherlands, he had a discussion with a well-known professor. Despite the reputation of his antagonist, Pierre firmly maintained his point of view, which has since been proven correct. He was also an excellent teacher, who had a particular concern for explaining to nonspecialists or beginners the latest developments in statistical mechanics. His enthusiasm in tackling a physics problem was stimulating to all who had the opportunity to work with him. In this note we stress the main aspects of his work.

1. THE GENERALIZED KINETIC EQUATIONS

Résibois' early work is concerned with the attempt initiated around 1950 to generalize the Boltzmann equation, which had been for almost 100 years the keystone of nonequilibrium physics. An important breakthrough occurred with the pioneering work of Bogolubov⁽¹⁾ and mostly van Hove⁽²⁾ and Prigogine and co-workers.⁽³⁾ His major contribution in this field was the derivation in collaboration with Prigogine of a general master equation for the N -particle velocity distribution function $\rho_N(v, t)$,^[17]

$$\partial_t \rho_N(v, t) = \int_0^t dr G_N(t-r) \rho_N(r) + D_N(v, t) \quad (1)$$

where $G_N(r)$ is a non-Markovian kernel, and $D_N(t)$ is a term describing the destruction of the initial correlations; a creation operator is also introduced which corresponds to the creation of fresh correlations by the mechanical interactions. In the long-time limit, Eq. (1) has been shown to reduce to the following Markovian equation^[17]:

$$\partial_t \rho_N(v, t) = \Theta \rho_N(v, t); \quad \Theta = \Omega \psi(i0) \quad (2)$$

Here $\psi(z)$ is the Laplace transform of the kernel appearing in Eq. (1) and Ω is a functional of ψ . This equation has played an important role in the subsequent development of this theory, which offers a description of the evolution of the system in terms of dynamics of correlations. Résibois also transposed the whole formalism to the quantum mechanical case.^[16] He then made a detailed comparison of the various approaches mentioned above.^[27] In the quantum theory, the time ordering of the various elementary processes allows one to define in a unique way a collision process characterized by the collision operator $\psi(z)$.^[39] Résibois performed an

extensive analysis of the structure of the three-body collision operator.^[41,42,51]

He also undertook a study of the transport phenomena in dense inhomogeneous systems starting from the linear generalized Boltzmann equation

$$\partial_t f_q(v, t) + i\mathbf{q} \cdot \mathbf{v} f_q(v, t) = \Omega_q \psi_q f_q(v, t) \quad (3)$$

$f_q(v, t)$ is the Fourier transform of the one-particle distribution function (d.f.). He first demonstrated the complete equivalence between the kinetic approach based on Eq. (3) and the correlation function formalism.^[37] He then applied the theory to the study of the transport phenomena in electrolytes by taking into account the collective effects due to the long-range character of the Coulomb forces.^[32,45,60] In collaboration with H. T. Davis and J. L. Lebowitz he developed in this formalism a microscopic theory of Brownian motion based on an $(m/M)^{1/2}$ expansion of transport properties of a large, heavy particle of mass M in a fluid of light particles of mass m .^[34,44] An extension of this theory to quantum fluids^[52,53] gives a satisfactory description of the motion of an ion in ${}^4\text{He}$.⁽⁴⁾ His book, of a mainly pedagogic character, *Theory of Electrolytes: An Elementary Microscopic Approach*, illustrates the application of statistical mechanics to the study of the equilibrium and nonequilibrium properties of electrolytes.

2. IRREVERSIBILITY IN LOCALIZED SPIN SYSTEMS

One of the most original of Résibois' contributions to nonequilibrium statistical mechanics is provided by his analysis of irreversibility in localized spin systems. The prototype of such systems is given by the isotropic Heisenberg model characterized by the following Hamiltonian:

$$H = - \sum_{i>j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j$$

J_{ij} is the exchange integral between the spins at lattice points i and j . The study of the dissipative properties of these models offers a challenging problem. Indeed, the Hamiltonian cannot be split into an unperturbed part plus a perturbation; there is a single time scale. As a result the traditional methods based on a generalization of Boltzmann's ideas are not applicable.

From the experimental point of view, the spin autocorrelation function $\Gamma_{ab}(t)$,

$$\Gamma_{ab}(t) = \langle S_a(t) S_b(0) \rangle \quad (4)$$

has been carefully analyzed by neutron scattering experiments and nuclear magnetic resonance.

In collaboration with M. De Leener, Résibois in 1966 derived kinetic equations for the correlation functions of the Heisenberg model in the high-temperature limit.^[54,55] They take the form

$$\begin{aligned}\partial_t \Gamma_q(t) &= \int_0^t G_q(r) \Gamma_q(t-r) dr \\ \Gamma_q(t) &= \sum_b \{ \exp[iq(\mathbf{ab})] \} \Gamma_{ab}(t)\end{aligned}\quad (5)$$

The whole problem consisted in finding a suitable explicit approximation for $G_q(r)$. The collision process between two localized spins being arbitrarily long, one is not allowed to consider these spins as isolated. One has to take into account that the other spins of the system play the role of a bath which dissipates the magnetization put initially on the given spins. This dissipation is described by the direct autocorrelation function $\Gamma(r)$. This qualitative argument was nicely formalized in theorems by Résibois and De Leener (RDL) in the Weiss limit where the number of neighbors goes to infinity. In this limit the perturbation series for $G_q(r)$ can be rewritten as a nonlinear functional of $\Gamma(r)$. A systematic approximation scheme was then proposed. In subsequent papers they extended this theory at finite temperatures (above the critical point) by including the equilibrium correlations; the kernel then becomes nonlinear in the complete autocorrelation function $\Gamma_q(r)$.^[62,63] At small q and away from the critical region, this equation reduces for long times to the spin diffusion equation, and an explicit expression for the diffusion coefficient is obtained.^[55] The extension of this theory to nuclear spin systems⁽⁵⁾ has allowed the interpretation of various experiments, such as, for instance, the long-time exponential behavior of the high magnetic field induction decay.^[56]

The equations were then used to study the critical fluctuations in ferromagnetic and antiferromagnetic systems.^[57] Close to $T_c(T_n)$ the solutions satisfy the dynamical scaling laws (DSA),⁽⁶⁾ and the characteristic frequency of these fluctuations may be written

$$\begin{aligned}\omega_q(\kappa) &= q^{5/2} f(q/\kappa) && \text{for ferromagnets} \\ \omega_q^A(\kappa) &= q^{3/2} f_A(q/\kappa) && \text{for staggered magnetization in antiferromagnets}\end{aligned}$$

Similar results have been obtained by the simpler mode-mode coupling method.⁽⁷⁾ However, this kinetic theory furnishes for a well-defined model a microscopic justification of the DSA and the mode-mode approach, which are usually based on phenomenological or macroscopic considerations. With Hohenberg, RDL also presented a detailed comparison between the DSA and the microscopic description for arbitrary dimension d .^[89] Résibois and Piette^[72,90] calculated explicitly the homogeneous functions $f(x)$ and $f_A(x)$, which remain unspecified in the DSA. In the case of

antiferromagnets, they obtained a good agreement with the experiments on RbMnF_3 .^(8,9) For ferromagnets they made predictions which have since been experimentally tested.⁽¹⁰⁾ The theory was then extended below the critical point in order to describe the dynamical behavior of the system in all the ordered region.^[80,83] The method has also been applied to the study of dynamic correlation functions of H-bonded ferroelectrics.^[75] A detailed numerical analysis of these equations and a comparison with experiments have been presented recently.⁽¹¹⁾ An extension of the theory to anisotropic systems has allowed the description of the crossover from isotropic to uniaxial behavior in reasonable agreement with experiments on FeF_2 and MnF_2 .⁽¹²⁾

3. HYDRODYNAMIC MODES IN STATISTICAL PHYSICS

The concept of hydrodynamic modes plays an important role in the many problems of statistical physics where long-wavelength, low-frequency behavior dominates, such as, for instance, in the critical behavior of transport coefficients⁽¹³⁾ or the slow power-law decay of correlation functions in two and three dimensions.⁽¹⁴⁾ In 1969, Résibois formulated the problem of the linear hydrodynamic modes from a microscopic point of view.^[69] He treated the eigenvalue problem associated to the linear generalized Boltzmann equation [Eq. (3)] by a degenerate perturbation method in powers of the uniformity parameter q . The hydrodynamic frequencies appear then as the five eigenvalues calculated up to the order q^2 which tend to zero when $q \rightarrow 0$. As a by-product, transport coefficients including their potential part can be computed, and the explicit equivalence with the autocorrelation formulas has been proved.^[70] This analysis clarifies the microscopic interpretation of transport coefficients with respect to the traditional approach, which was based on a generalization to dense systems of the method introduced by Chapman and Enskog for dilute gases. On the other hand, the eigenfunctions associated to the hydrodynamic frequencies furnish a one-body microscopic expression for the hydrodynamic modes. This property proved useful in the treatment of a large variety of problems where the hydrodynamic limit of the propagators plays the dominant role:

1. The long-wavelength, low-frequency limit of the van Hove correlation function.^[71]
2. The kinetic energy density fluctuations in classical liquids.^[81]
3. The long-wavelength behavior of the Green–Kubo integrands.^[92–94]
4. Two-dimensional hydrodynamics.^[110]

The various approaches to the study of the long-wavelength hydrodynamical processes have been beautifully summarized by Résibois and Pomeau in a report which is now a classic reference for a review on the long-time behavior of the Green–Kubo integrands.^[98]

4. TRANSPORT PROPERTIES OF THE VAN DER WAALS FLUID

Another interesting application of Résibois' theory of hydrodynamic modes is furnished by the study of transport properties of van der Waals fluids. In collaboration with Piasecki and Pomeau, he performed a microscopic calculation of the transport coefficients of a system the interaction potential of which may be written^[84,88]

$$V(r) = V^S(r) + \gamma^3 V^L(r) \quad (6)$$

V^S refers to the short-range reference system, and V^L is the long-range part of the potential with range γ^{-1} . They analyzed the properties of this system in the limit $\gamma \rightarrow 0$. Starting from the Green-Kubo formula they developed a renormalized perturbation scheme; the linearized collision operator then becomes a functional of the propagators:

$$G_q(\mathbf{v}_1) = \lim_{\epsilon \rightarrow 0} (\epsilon - i\mathbf{q}\mathbf{v}_1 + i\psi'_q)^{-1} \quad (7)$$

$G_q(\mathbf{v}_1)$ describes the exact motion of particle 1 in the dense medium. They derived an expansion of the transport coefficient X in positive powers of γ :

$$X = X^S + \gamma \delta X_1 \quad (8)$$

An explicit evaluation of the first correction was obtained by applying a nontrivial generalization of the theory of hydrodynamic modes.^[88] A numerical computation of the first correction to the shear viscosity was performed.^[105] In the vicinity of the critical point, the theory provides a reasonable description of the onset of critical phenomena.^[105] This study adds a new system to the few solvable models in nonequilibrium statistical mechanics where the solvability is connected with the existence of a smallness parameter.

5. HARD-SPHERE DYNAMICS

Irreversibility in hard-sphere systems offers among the most challenging problems in statistical mechanics. Indeed, their properties can all be expressed in terms of a single parameter which is of the order of unity in dense systems. Different approaches have been developed to tackle this problem. Already in 1925, Enskog proposed a phenomenological equation which provides a surprisingly good description of the behavior of these systems even at fairly high densities.⁽¹⁵⁾ On the other hand, great progress has been achieved in the ergodic theory of these models.⁽¹⁶⁾ Résibois and Lebowitz proposed an approximate kinetic theory which offers a systematic procedure for improving the old Enskog theory.^[99] In a subsequent paper, Résibois applied this theory to the evaluation of the velocity correlation function at arbitrary times and densities.^[102] The results of this numerical

evaluation are quite satisfactory. For instance, the deviations from the Enskog value are well reproduced at all densities. Similarly, the values D/D_E (D being the diffusion coefficient) are very good, especially the strong decrease of this ratio at the highest densities.

More recently, Résibois demonstrated an H theorem for the modified Enskog equation.^{(17),[114,116]} He constructed a functional S of the one-particle distribution function f_1 which reduces at equilibrium to the exact entropy. Assuming periodic boundary conditions, he proved that $\partial S/\partial t \geq 0$. He also showed that S tends to a stationary value when f_1 is the absolute equilibrium d.f. Work along these lines is being pursued by Mareschal.⁽¹⁸⁾

In collaboration with De Leener, Résibois wrote a coherent and self-contained introduction to the nonequilibrium theory of classical fluids, *Classical Kinetic Theory of Fluids*.

As is nicely illustrated in the present collection of papers, the study of irreversible processes has attracted many people from various fields ranging from macroscopic to microscopic physics. However, more than 100 years after the publication of Boltzmann's paper, the formulation of a consistent microscopic theory of irreversibility remains a basic problem of theoretical physics. In this context, Résibois' work has had a great importance because it has led to a better formalization of the problem on a purely technical basis. In order to appreciate the contribution of Résibois in this domain, it is important to remember that for a long time kinetic equations were derived only for specific limiting cases and they were based at least partly on phenomenological arguments. Even today in many fields of nonequilibrium statistical mechanics ad hoc approximations are introduced in an uncontrollable fashion. It is difficult to believe that the emergence of concepts such as, for instance, a microscopic definition of entropy could ever be explained by some form of coarse-graining of a finer, exact description. We think that new mathematical tools and new concepts have to be introduced in order to construct a proper framework for a microscopic theory of irreversible processes. In this perspective the work of Pierre Résibois occupies an everlasting position.

LIST OF PUBLICATIONS OF PIERRE RÉSIBOIS

A. Books

- P. Résibois, *Theory of Electrolytes: An Elementary Microscopic Approach* (Harper and Row, New York, 1967).
 P. Résibois and M. De Leener, *Classical Kinetic Theory of Fluids* (Wiley, New York, 1977).

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