Proceedings of the XXIth Solvay Conference on Physics "Dynamical Systems and Irreversibility", Japan, November 1-5, 1998;
Advances in Chemical Physics **122** (2002) 109-127.

Diffusion and the Poincaré-Birkhoff mapping of chaotic systems

Pierre GASPARD
Center for Nonlinear Phenomena and Complex Systems,
Université Libre de Bruxelles,
Campus Plaine C. P. 231, Blvd du Triomphe,
B-1050 Brussels, Belgium

I. INTRODUCTION

In the present contribution, we summarize recent work on diffusion in spatially periodic chaotic systems. The hydrodynamic modes of diffusion are constructed as the eigenstates of a Frobenius-Perron operator associated with the Poincaré-Birkhoff mapping of the system. The continuous-time dynamics is included in the Frobenius-Perron operator of the mapping by using the first-return time function. We show that the transport properties of diffusion can be derived from this Frobenius-Perron operator, such as the Green-Kubo relation for the diffusion coefficient, the Burnett and higher-order coefficients, the Lebowitz-McLennan-Zubarev expression for the nonequilibrium steady states, and the entropy production of nonequilibrium thermodynamics. The hydrodynamic modes are given by conditionally invariant complex measures with a fractal-like singular character. The construction of the hydrodynamic modes is inspired of works by Prigogine [1], Balescu [2], Résibois [3], Boon [4], and others.

Recent progress in statistical mechanics has revealed the importance of chaos to understand normal transport processes such as diffusion, viscosity or heat conductivity. During this last decade, many works have shown that microscopic systems of interacting particles are typically chaotic in the sense that their dynamics is highly sensitive to initial conditions because of positive Lyapunov exponents and that this dynamical instability leads to a dynamical randomness characterized by a positive Kolmogorov-Sinai (KS) entropy per unit time [5–12]. This dynamical randomness can induce various transport phenomena, which turn out to be *normal* in the sense that the associated fluctuations are of Gaussian character on long times and that the associated hydrodynamic modes decay exponentially.

Such results have been obtained for systems with few mutually interacting degrees of freedom. In particular, diffusion has been extensively studied in systems such as the Lorentz gases, in which a point particle undergoes elastic collisions in a periodic lattice of ions. The ions have been modeled as hard disks [13, 14] or as centers of Yukawa-type potentials [15]. Normal diffusion has also been studied in simplified models such as the multibaker maps [16–19]. In the same context, Gaussian thermostated Lorentz gases have also been studied [20, 21]. In these thermostated systems, the particle is submitted to an external field and to a deterministic force which is nonHamiltonian. As a consequence, phase-space volumes are no longer preserved in thermostated systems, which constitutes a fundamental difficulty of this approach. Non-area-preserving multibaker maps have also been considered [22]. In this case, an equivalence was shown between the non-area-preserving maps and area-preserving maps with an extra variable of energy allowing the phase space to be larger as the particle gains kinetic energy [23, 24]. In this contribution, we shall focus on transport in systems which preserve phase-space volumes so that the sum of all the Lyapunov exponents vanishes $\sum_i(\lambda_i^+ + \lambda_i^-) = 0$, in agreement with Liouville's theorem of Hamiltonian mechanics. For systems of statistical mechanics, Liouville's theorem is an important property because it is a consequence of the unitarity of the underlying quantum mechanics ruling the microscopic dynamics.

The purpose of the present contribution is to give a short synthesis of the studies of diffusion and other transport processes in spatially periodic chaotic systems which obeys Liouville's theorem. In these studies, a special role is played by the Poincaré-Birkhoff mapping, which provides a powerful method to reduce the continuous-time evolution of statistical ensembles of trajectories to a discrete-time Frobenius-Perron operator [25]. The concept of microscopic chaos and the escape-rate formalism will also be shortly reviewed.

The contribution is organized as follows. Section 2 contains a discussion about chaos in nonequilibrium statistical mechanics and about the escape-rate formalism. In Sec. 3, we show how a flow can be reduced to a Poincaré-Birkhoff mapping and similarly how, in spatially periodic systems, Liouville's equation can be studied in terms of a special Frobenius-Perron operator of our invention. In Sec. 4, we show that all the known formulae of the transport theory of diffusion can be derived from our Frobenius-Perron operator and its classical Pollicott-Ruelle resonances. Applications and conclusions are given in Sec. 5.

II. MICROSCOPIC CHAOS

In chaotic systems, the sensitivity to initial conditions generates a time horizon which is of the order of the inverse of the maximum Lyapunov exponent $\lambda_{\max}^+ = \max\{\lambda_i^+\}$, multiplied by the logarithm of the ratio of the final error $\varepsilon_{\rm final}$ over the initial one $\varepsilon_{\rm initial}$:

$$t_{\rm Lyapunov} \; = \; \frac{1}{\lambda_{\max}^+} \; \ln \frac{\varepsilon_{\rm final}}{\varepsilon_{\rm initial}} \; . \tag{1}$$

This Lyapunov time constitutes a horizon for the prediction of the future trajectory of the system [26]. Starting from initial conditions known to a precision given by $\varepsilon_{\rm initial}$, the trajectory keeps a tolerable precision lower than $\varepsilon_{\rm final}$ only during the time interval $0 \leq t < t_{\rm Lyapunov}$. We notice that the Lyapunov horizon is movable in the sense that it can be extended to a longer time by decreasing the initial error $\varepsilon_{\rm initial}$. Nevertheless, this requires to increase exponentially the precision on the initial conditions.

In a dilute gas of interacting particles, a positive Lyapunov exponent has the typical value [5, 6]

$$\lambda_i^+ \; \sim \; \frac{v}{\ell} \; \ln \frac{\ell}{d} \; , \tag{2}$$

where d is the particle diameter, $v \sim \sqrt{k_{\rm B}T/m}$ is the mean velocity, and $\ell \sim (nd^2)^{-1}$ is the mean free path given in terms of the particle density n. At room temperature and pressure, a typical Lyapunov exponent takes the value $\lambda_i^+ \sim 10^{10}$ sec^{-1}, which is of the order of the inverse of the intercollisional time. There are as many positive Lyapunov exponents as there are unstable directions in phase space. Pesin's formula [27] can be used to evaluate the KS entropy per unit time of a mole of particles at equilibrium

$$h_{\rm KS} = \sum_i \lambda_i^+ \; \sim \; 3 \; N_{\rm Av} \; \frac{v}{\ell} \; \ln \frac{\ell}{d} \; , \tag{3}$$

where $N_{\rm Av}$ is Avogadro's number. The KS entropy characterizes the dynamical randomness of the motion of the particles composing the gas. The effect of this microscopic chaos has been observed in a recent experiment where a positive lower bound has been measured on the KS entropy per unit time of a fluid containing Brownian particles [28].

At equilibrium, the formula (3) shows that the dynamical instability is converted into an exactly equal amount of dynamical randomness. The reason for this is that the dynamical instability cannot proceed forever in nonlinear Hamiltonian systems where the explosion is stopped by nonlinear saturations. In this way, dynamical randomness is generated by some mechanisms of nonlinear saturation during the dynamics. The exact compensation of the dynamical instability by the dynamical randomness is an interesting feature expressed by Pesin's formula (3).

In nonequilibrium situations, this exact compensation is broken leading to the generation of fractal structures in the phase space of the system of particles [11, 29–32]. In the escape-rate formalism, a nonequilibrium situation is induced by absorbing boundaries in the phase space. These absorbing boundaries describe for instance an experiment in which the system is observed until a certain property reaches a certain threshold or exits a certain range of values or a domain of motion. The time of this event is recorded and the experiment is restarted. After many repetitions, a statistics of first-passage times can be performed. Very often, these first-passage times are distributed exponentially, which defines a rate of exponential decay called the *escape rate*. This rate depends on the observed property as well as on the chosen threshold, i.e., on the geometry of the absorbing boundaries. If the observed property is the position of a tracer particle diffusing in a fluid, the escape rate turns out to be proportional to the diffusion coefficient. Similarly, if the observed property is the center of momentum of all the particles composing the fluid, the escape rate is proportional to the viscosity, etc... [32].

At the microscopic level, the absorbing boundaries select highly unstable trajectories along which the observed property never reaches the threshold. In chaotic systems, infinitely many such trajectories exist in spite of the fact that most trajectories reach the threshold and are absorbed at the boundary (or, equivalently, they escape out of the domain delimited by the boundaries). Accordingly, all these trajectories which evolve forever inside the boundaries without reaching them form a so-called *fractal repeller*. On this fractal repeller, the KS entropy is slightly smaller than the sum of positive Lyapunov exponents and the difference is equal to the escape rate [27]

$$\gamma \; = \; \sum_i \lambda_i^+ \; - \; h_{\rm KS} \; . \tag{4}$$

Since the escape rate γ depends on the transport coefficient, it is possible to relate the transport coefficients to the characteristic quantities of chaos, as shown elsewhere [11, 30–32].

A priori, it is not evident that such a relationship is possible because the microscopic chaos is a property of the short time scale $(\lambda_i^+)^{-1} \sim 10^{-10}$ sec of the collisions between the particles although the transport properties manifest themselves on the long hydrodynamic time scale. However, the nonequilibrium conditions created by the absorbing boundaries lead to a small difference between the KS entropy and the sum of positive Lyapunov exponents. This difference is of the order of the inverse of a hydrodynamic time such as the diffusion time for a tracer particle to reach the absorbing boundaries. Thanks to the escape-rate formalism [11, 30–32], the connection between both types of properties thus becomes possible because a difference is taken between two properties from the short intercollisional time scale.

Subsequent work has shown the fundamental importance of the fractal repeller of the escape-rate formalism [11, 19, 25, 33, 34]. This fractal repeller is closely associated with the other fractals which appear in infinitely extended systems without absorbing boundaries. For instance, it has been possible to show that, in infinitely extended systems such as the periodic Lorentz gas, the nonequilibrium steady states of diffusion are described by singular measures (see Sec. 4) [19, 25]. The singularities of these measures originate from the discontinuities of the invariant density in a large but finite system between two reservoirs of particles [33]. These discontinuities occur on the unstable manifolds of the fractal repeller of the trajectories evolving forever between the two reservoirs. In turn, the singular measures describing the nonequilibrium steady states are the derivatives of the diffusive hydrodynamic modes with respect to their wavenumber. As a consequence, these hydrodynamic modes are also described by singular measures with their singularities intimately related to the same fractal repeller of the escape-rate formalism. Since the singular character of these measures is at the origin of the positive entropy production of nonequilibrium thermodynamics (as shown in Sec. 4) [34], it turns out that the fractal repeller of the escape-rate formalism plays a basic and fundamental role in this whole context. Let us here mention that the escape-rate formalism is closely related to the scattering theory of transport by Lax and Phillips [35]. These authors have pointed out the importance of the set of trapped trajectories – today called the fractal repeller – and of its stable and unstable manifolds in order to define a classical scattering operator.

III. SPATIALLY PERIODIC SYSTEMS

A. Poincaré-Birkhoff mapping in spatially periodic systems

We consider a continuous-time system with a phase space which is periodic in some directions. This is the case for the periodic Lorentz gas in which a point particle undergoes elastic collisions in a lattice of hard disks or interacts with a lattice of Yukawa attracting potentials. In the first example [13, 14], the system is defined by the Hamiltonian of kinetic energy for the free motion between the hard disks and by the condition that the position (x, y) remains outside the hard disks of diameter d forming a regular lattice $\mathcal{L}$:

$$H = \frac{p_x^2 + p_y^2}{2m} \ , \tag{5}$$

$$\sqrt{(x-l_x)^2 + (y-l_y)^2} \geq d/2 \ , \qquad \text{for} \quad \mathbf{l} = (l_x, l_y) \in \mathcal{L} \ . \tag{6}$$

In the second example [15], the Hamiltonian is given by

$$H = \frac{p_x^2 + p_y^2}{2m} + \sum_{\mathbf{l}\in\mathcal{L}} V(\mathbf{r}-\mathbf{l}) \ , \qquad \text{with} \quad V(r) = -\frac{\exp(-\alpha r)}{r} \ . \tag{7}$$

Another example is a system composed of a tracer particle moving among $(N-1)$ other particles with periodic boundary conditions, as usually assumed in numerical simulations.

Such systems are described by some differential equations

$$\frac{d\mathbf{X}}{dt} = \mathbf{F}(\mathbf{X}) \ , \tag{8}$$

for M variables $\mathbf{X}$. If the conditions of Cauchy's theorem are satisfied, the trajectory at time t is uniquely given in terms of the initial conditions $\mathbf{X}_0$, which defines the continuous-time flow:

$$\mathbf{X}_t = \mathbf{\Phi}^t(\mathbf{X}_0) \ . \tag{9}$$

Because of the periodicity, the phase space can be divided into a lattice of cells within each of them the vector field (8) is the same. One of these cells defines the fundamental cell of our lattice in the phase space. The flow in the full phase space can be reduced to the flow in the fundamental cell with periodic boundary conditions. The motion on the lattice of cells is followed by a vector $\mathbf{l}$ belonging to the lattice $\mathcal{L}$. A Poincaré surface of section $\mathcal{P}$ can be defined in this fundamental cell of the phase space. This surface of section is equipped with $M-1$ coordinates $\mathbf{x}$. During the time evolution, a trajectory (9) will intersect the surface of section successively at the points $\{\mathbf{x}_n\}_{n=-\infty}^{+\infty}$ and the times $\{t_n\}_{n=-\infty}^{+\infty}$. At each intersection with the surface of section, the vector locating the trajectory in the lattice is updated so that a sequence of lattice vectors $\{\mathbf{l}_n\}_{n=-\infty}^{+\infty}$ is furthermore generated by the motion.

Because the system is deterministic, each intersection $\mathbf{x}$ uniquely determines the next intersection by a nonlinear map $\boldsymbol{\varphi}(\mathbf{x})$ called the Poincaré-Birkhoff mapping, a time of first return $T(\mathbf{x})$, and a vector-valued function $\mathbf{a}(\mathbf{x}) \in \mathcal{L}$ giving the jump on the lattice [25]:

$$\begin{cases} \mathbf{x}_{n+1} = \boldsymbol{\varphi}(\mathbf{x}_n) \ , \\ t_{n+1} = t_n + T(\mathbf{x}_n) \ , \\ \mathbf{l}_{n+1} = \mathbf{l}_n + \mathbf{a}(\mathbf{x}_n) \ . \end{cases} \tag{10}$$

Reciprocally, the flow can be expressed as a suspended flow in terms of the quantities introduced in the construction of the Poincaré-Birkhoff mapping. In order to recover the M coordinates $\mathbf{X}$ of the original flow, we need to introduce an extra variable τ beside the $M-1$ coordinates $\mathbf{x}$ of the Poincaré surface of section. The extra variable τ is the time of flight since the last intersection with the surface of section. This variable ranges between zero and the time $T(\mathbf{x})$ of first return in the section: $0 \leq \tau < T(\mathbf{x})$. Thanks to these coordinates, the suspended flow takes an explicit form given by [25]

$$\tilde{\boldsymbol{\Phi}}^t(\mathbf{x},\tau,\mathbf{l}) = \left[\boldsymbol{\varphi}^n\mathbf{x}, \tau + t - \sum_{j=0}^{n-1} T(\boldsymbol{\varphi}^j\mathbf{x}), \mathbf{l} + \sum_{j=0}^{n-1} \mathbf{a}(\boldsymbol{\varphi}^j\mathbf{x})\right] \ , \tag{11}$$

for $0 \leq \tau + t - \sum_{j=0}^{n-1} T(\boldsymbol{\varphi}^j\mathbf{x}) < T(\boldsymbol{\varphi}^n\mathbf{x})$. According to Cauchy's theorem and the geometric construction, there exists a function $\mathbf{G}$ which connects the variables of the suspended flow to the original ones:

$$\mathbf{X} = \mathbf{G}(\mathbf{x},\tau,\mathbf{l}) \qquad \text{with} \quad \mathbf{x} \in \mathcal{P}, \quad 0 \leq \tau < T(\mathbf{x}), \quad \mathbf{l} \in \mathcal{L} \ , \tag{12}$$

and an isomorphism is established in this way between the suspended and the original flows:

$$\tilde{\boldsymbol{\Phi}}^t = \mathbf{G}^{-1} \circ \boldsymbol{\Phi}^t \circ \mathbf{G} \ . \tag{13}$$

We notice that the jump vector is given by

$$\mathbf{a}(\mathbf{x}) \ = \ \mathbf{r}(\mathbf{x}) + \int_0^{T(\mathbf{x})} \mathbf{v} \circ \boldsymbol{\Phi}^\tau \circ \mathbf{G}(\mathbf{x},0,\mathbf{0}) \, d\tau \ - \ \mathbf{r}(\boldsymbol{\varphi}\mathbf{x}) \ , \tag{14}$$

in terms of the velocity $\mathbf{v}$ of the tracer particle at the current position in the phase space, if $\mathbf{r}(\mathbf{x})$ denotes the position with respect to the center of the fundamental cell at the instant of the intersection $\mathbf{x}$ with the surface of section.

B. Frobenius-Perron operator

Beyond the Lyapunov horizon, the predictability on the future trajectory is lost and statistical statements become necessary. Since the relaxation toward the thermodynamic equilibrium is a process taking place on asymptotically long times its description requires the introduction of statistical ensembles of trajectories.

In a multi-particle system, the use of statistical ensembles corresponds to the repetition of the same experiment over and over again until the statistical property is established with confidence. This is the case in scattering experiments for instance in the study of atomic or molecular collisions. Two beams of particles are sent onto each other and the scattering cross-section of elastic or inelastic collisions can be measured. These cross-sections are such statistical properties. In a scattering experiment, each beam contains an arbitrarily large number of particles arriving one after the other with statistically distributed velocities, impact parameters and arrival times. In a beam, the particles are sufficiently separated to be considered as independent so that the experiment can be described as a succession of independent binary collisions, forming a statistical ensemble of events.

In a single-particle system such as the Lorentz gas, the statistical ensembles are introduced for the same use as aforementioned. In such systems, the statistical ensemble can also be considered for the description of a gas of independent particles bouncing in a lattice of ions. Indeed, a statistical ensemble of N trajectories of the bouncing particle is strictly equivalent to a gas of N independent particles bouncing in the system. In the limit $N \to \infty$, the phase-space distribution of the statistical ensemble is equivalent to the position-velocity distribution of the gas of independent particles. The relaxation toward the equilibrium of the former is thus equivalent to the relaxation of the latter. Our purpose is here to describe this relaxation at the microscopic level without approximation.

The statistical ensemble is described by a probability density $\rho(\mathbf{X})$ defined in the phase space. The time evolution (8)-(9) of each trajectory induces a time evolution for the whole statistical ensemble according to Liouville's equation which takes one of the following equivalent forms [1, 26]:

$$\partial_t \, \rho \;=\; -\,\mathrm{div}(\mathbf{F}\rho) \;=\; \{H,\rho\} \;\equiv\; \hat{L}\,\rho \; , \tag{15}$$

where $\mathbf{F}$ is the vector field (8), H is the Hamiltonian, $\{\cdot,\cdot\}$ is the Poisson bracket, and $\hat{L}$ is the so-defined Liouvillian operator. As aforementioned, Liouville's equation (15) also rules the position-velocity density of a Lorentz gas of independent particles.

In the same way as the equations of motion (8) can be integrated to give the flow (9), the time integral of Liouville's equation gives the probability density ρ_t at the current time t in terms of the initial probability density ρ_0 according to

$$\rho_t(\mathbf{X}) \;=\; \exp(\hat{L}t)\,\rho_0(\mathbf{X}) \;=\; \rho_0(\mathbf{\Phi}^{-t}\mathbf{X}) \;\equiv\; \hat{P}^t\rho_0(\mathbf{X}) \; , \tag{16}$$

which defines the Frobenius-Perron operator, here given for volume-preserving systems.

In Ref. [25], we have shown how this continuous-time Frobenius-Perron operator decomposes into the discrete-time Frobenius-Perron operator associated with the Poincaré-Birkhoff mapping (10), which we assume to be area-preserving. This reduction is carried out by a Laplace transform in time which introduces the rate variable s conjugated to the time t. Furthermore, since the system is spatially periodic, we perform a spatial Fourier transform which introduces the wavenumber $\mathbf{k}$. In this way, the probability density decomposes into the so-called hydrodynamic modes of diffusion. Each of them corresponds to spatially quasiperiodic inhomogeneities of wavelength $L = 2\pi/\|\mathbf{k}\|$ in the phase-space density. Accordingly, the Frobenius-Perron operator decomposes into a continuum of Frobenius-Perron operators, one for each wavenumber $\mathbf{k}$ [25]:

$$(\hat{R}_{\mathbf{k},s}f)(\mathbf{x}) = \exp\left[-sT(\boldsymbol{\varphi}^{-1}\mathbf{x}) - i\,\mathbf{k}\cdot\mathbf{a}(\boldsymbol{\varphi}^{-1}\mathbf{x})\right]\; f(\boldsymbol{\varphi}^{-1}\mathbf{x}) \; , \tag{17}$$

where $f(\mathbf{x})$ is the $\mathbf{k}$-component of the density defined in the Poincaré surface of section $\mathcal{P}$. For $\mathbf{k}=0$, the Frobenius-Perron operator (17) reduces to an operator previously derived by Pollicott [36, 37].

In the Frobenius-Perron operator (17), the Laplace transform in time has introduced an exponential factor $\exp\left[-sT(\boldsymbol{\varphi}^{-1}\mathbf{x})\right]$ where $T(\boldsymbol{\varphi}^{-1}\mathbf{x})$ is the time of first return in the surface of section $\mathcal{P}$ after a previous intersection at $\boldsymbol{\varphi}^{-1}\mathbf{x}$. This factor has the following interpretation. Let us suppose that the Frobenius-Perron operator rules the time evolution of a mode with a decay rate $\gamma = -s$ such that $\mathrm{Re}\,\gamma > 0$. The operator (17) should describe locally in the phase space the time evolution from the previous intersection $\boldsymbol{\varphi}^{-1}\mathbf{x}$ up to the current intersection $\mathbf{x}$ with $\mathcal{P}$. During the first-return time $T(\boldsymbol{\varphi}^{-1}\mathbf{x})$, the density f decays exponentially by an amount $\exp\left[-\gamma T(\boldsymbol{\varphi}^{-1}\mathbf{x})\right]$. The first factor in Eq. (17) is there to compensate this decay in order to define the conditionally invariant density associated with the decay mode.

A similar interpretation holds for the factor $\exp\left[-i\mathbf{k}\cdot\mathbf{a}(\boldsymbol{\varphi}^{-1}\mathbf{x})\right]$. During the same segment of trajectory from $\boldsymbol{\varphi}^{-1}\mathbf{x} \in \mathcal{P}$ to $\mathbf{x} \in \mathcal{P}$, the particle has moved in the lattice by a vector $\mathbf{a}(\boldsymbol{\varphi}^{-1}\mathbf{x})$ so that the Fourier $\mathbf{k}$-component f acquires the phase $\exp\left[+i\mathbf{k}\cdot\mathbf{a}(\boldsymbol{\varphi}^{-1}\mathbf{x})\right]$ during this motion. The second factor has thus the effect to compensate this phase in order for the component f to continue to describe the density in the fundamental cell.

According to the previous discussion, we can conclude that a hydrodynamic mode of wavenumber $\mathbf{k}$ will be given by a solution of the generalized eigenvalue problem [25]:

$$\hat{R}_{\mathbf{k},s_{\mathbf{k}}}\,\psi_{\mathbf{k}} \;=\; \psi_{\mathbf{k}} \; , \tag{18}$$

$$\hat{R}^{\dagger}_{\mathbf{k},s_{\mathbf{k}}}\,\tilde{\psi}_{\mathbf{k}} \;=\; \tilde{\psi}_{\mathbf{k}} \; , \tag{19}$$

with the bi-orthonormality condition taken with respect to the invariant Lebesgue measure ν defined in the surface of section:

$$\langle\tilde{\psi}^{*}_{\mathbf{k}}\;\psi_{\mathbf{k}}\rangle_{\nu} = 1 \; . \tag{20}$$

In Eqs. (18)-(20), $\psi_{\mathbf{k}}$ is the eigenstate of the Frobenius-Perron operator (17) and $\tilde{\psi}_{\mathbf{k}}$ is the adjoint eigenstate. In Eq. (18), the eigenvalue of the operator (17) is taken to be equal to unity in order for $\psi_{\mathbf{k}}$ to become the density associated with a conditionally invariant measure, as discussed above. Indeed, it has been necessary to include already the decay rate ($-s$) in the Frobenius-Perron operator (17) because the Poincaré-Birkhoff mapping (10) is not isochronic. Therefore, requiring that the eigenvalue of the operator (17) is equal to unity has the effect of fixing the variable s to a value which depends on the wavenumber $\mathbf{k}$ and which is intrinsic to the system. This value $s_{\mathbf{k}}$ is a so-called Pollicott-Ruelle resonance [36–41]. The Pollicott-Ruelle resonances have been theoretically studied in different systems such as the disk scatterers [42] or the pitchfork bifurcation [43] and, notably, in connection with relaxation and diffusion in classically chaotic quantum systems [44–49]. These resonances have also been evidenced in an experimental microwave study of disk scatterers [50, 51].

Several such Pollicott-Ruelle resonances may exist. In ergodic and mixing spatially periodic systems, we may expect that there is a unique Pollicott-Ruelle resonance which vanishes with the wavenumber: $\lim_{\mathbf{k}\to 0} s_{\mathbf{k}} = 0$. In this limit, the corresponding eigenstate becomes the invariant state which is the microcanonical equilibrium measure for volume-preserving Hamiltonian systems: $\lim_{\mathbf{k}\to 0} \psi_{\mathbf{k}} = 0$. However, for small enough nonvanishing wavenumbers $\mathbf{k} \neq 0$, the eigenstate is no longer the invariant measure and instead defines a complex conditionally invariant measure describing a hydrodynamic mode of diffusion.

In chaotic systems, these conditionally invariant measures are singular with respect to the Lebesgue measure so that their density $\psi_{\mathbf{k}}$ is not a function but a mathematical distribution (also called a generalized function) of a type defined by Schwartz or Gel'fand. A cumulative function can be defined as the measure of a m-dimensional rectangular domain $[\mathbf{0}, \mathbf{x}]^m$ in phase space with $0 < m \leq M - 1$:

$$F_{\mathbf{k}}(\mathbf{x}) \; = \; \int_{[\mathbf{0},\mathbf{x}]^m} \psi_{\mathbf{k}}(\mathbf{x}') \; d\mathbf{x}' \; , \tag{21}$$

which is expected to be continuous but nondifferentiable for small enough wavenumbers $\mathbf{k} \neq 0$ [17, 18, 25, 52–54]. In Eq. (21), $d\mathbf{x}'$ defines the invariant Lebesgue measure ν in the surface of section $\mathcal{P}$. For $m = 1$, the cumulative functions (21) of the hydrodynamic modes of the multibaker maps and of the two-dimensional Lorentz gases form fractal curves in the complex plane. These fractal curves are characterized by a Hausdorff dimension which is given in terms of the diffusion coefficient and the Lyapunov exponent [55, 56].

In the next section, we shall present the consequences of these results on the transport properties of diffusion.

IV. CONSEQUENCES OF THE EIGENVALUE PROBLEM

The generalized eigenvalue problem presented in the previous section defines the hydrodynamic modes of diffusion at the microscopic level of description. As summarized here below, all the relevant properties of the transport by diffusion such as the diffusion coefficient, the higher-order Burnett and super-Burnett coefficients if they exist, the nonequilibrium steady states, Fick's law, and the entropy production of nonequilibrium thermodynamics can be derived from the Frobenius-Perron operator (17).

A. The diffusion coefficient and the higher-order coefficients

We suppose that the leading Pollicott-Ruelle resonance $s_{\mathbf{k}}$ is n-times differentiable near $\mathbf{k} = 0$: $s_{\mathbf{k}} \in \mathcal{C}^n$. Hence, the eigenvalue equations (18)-(20) can be differentiated successively with respect to the wavenumber: $\partial_{\mathbf{k}}$, $\partial^2_{\mathbf{k}}$, $\partial^3_{\mathbf{k}}$, $\partial^4_{\mathbf{k}}$,...,$\partial^n_{\mathbf{k}}$ [25].

In the absence of external field, there is no mean drift and the first derivatives give

$$\partial_{\mathbf{k}} s_0 \; = \; 0 \; , \tag{22}$$

$$\partial_{\mathbf{k}} \psi_0(\mathbf{x}) \; = \; -i \sum_{n=1}^{\infty} \mathbf{a}(\boldsymbol{\varphi}^{-n}\mathbf{x}) \; . \tag{23}$$

Proceeding in a similar way up to the second derivatives gives us the matrix of diffusion coefficients (if they exist) as:

$$D_{\alpha\beta} = -\frac{1}{2} \frac{\partial^2 s_0}{\partial k_\alpha \partial k_\beta} = \frac{1}{2\langle T \rangle_\nu} \sum_{n=-\infty}^{+\infty} \langle a_\alpha (a_\beta \circ \boldsymbol{\varphi}^n) \rangle_\nu \; . \tag{24}$$

Using Eq. (14) which relates the jump vector $\mathbf{a}(\mathbf{x})$ to the integral of the particle velocity $\mathbf{v}$, Eq. (24) implies the Green-Kubo relation:

$$D_{\alpha\beta} = \frac{1}{2} \int_{-\infty}^{+\infty} dt \; \langle v_\alpha (v_\beta \circ \mathbf{\Phi}^t) \rangle_\mu \; , \tag{25}$$

where μ is the microcanonical equilibrium measure in the original phase space.

Higher derivatives give expressions for the Burnett and super-Burnett ($B_{\alpha\beta\gamma\delta}$) coefficients if they exist [25]. These higher-order coefficients appear in the expansion of the dispersion relation of diffusion in powers of the wavenumber:

$$s_{\mathbf{k}} = -\sum_{\alpha\beta} D_{\alpha\beta}\, k_\alpha\, k_\beta + \sum_{\alpha\beta\gamma\delta} B_{\alpha\beta\gamma\delta}\, k_\alpha\, k_\beta\, k_\gamma\, k_\delta + \mathcal{O}(k^6)\ . \tag{26}$$

In a recent work [57, 58], Chernov and Dettmann proved the existence of these higher-order coefficients as well as the convergence of the expansion (26) for the periodic Lorentz gas with a finite horizon, on the basis of the formalism described here.

B. Periodic-orbit theory

The periodic-orbit theory of classical systems has been extensively developed since the pioneering work by Cvitanović [59]. This theory has been applied to many dynamical systems which are either classical, stochastic, or quantum. For an overview of periodic-orbit theory, see Ref. [60].

For Axiom-A spatially periodic systems, the Fredholm determinant of the Frobenius-Perron operator (17) can be expressed as a product over all the unstable periodic orbits given by the following Zeta function [25]:

$$\begin{aligned} Z(s;\mathbf{k}) &\equiv \mathrm{Det}\big(\hat{I} - \hat{R}_{\mathbf{k},s}\big) \\ &= \prod_p \prod_{m_1\cdots m_u=0}^{\infty} \left[1 - \frac{\exp(-s\,T_p - i\,\mathbf{k}\cdot\mathbf{a}_p)}{|\Lambda_{1p}\cdots\Lambda_{up}|\Lambda_{1p}^{m_1}\cdots\Lambda_{up}^{m_u}}\right]^{(m_1+1)\cdots(m_u+1)}\ , \end{aligned} \tag{27}$$

where Λ_{ip} with $i = 1, ..., u$ are the instability eigenvalues of the linearized Poincaré-Birkhoff mapping (10) for the periodic orbit p. These instability eigenvalues satisfy $|\Lambda_{ip}| > 1$. T_p is the period of p and $\mathbf{a}_p$ is the vector by which the particle travels on the lattice during the period. The integer u is the number of unstable directions in the phase space.

According to the eigenvalue equation (18), the Pollicott-Ruelle resonances $s_{\mathbf{k}}$ are the zeroes of the Fredholm determinant: $Z(s_{\mathbf{k}};\mathbf{k}) = 0$. This result can be used in order to obtain a periodic-orbit formula for the diffusion coefficient of an isotropic d-dimensional diffusive process [61–63]:

$$D = \frac{\displaystyle\sum_{l=0}^{\infty}(-)^l \sum_{p_1\neq\cdots\neq p_l} (\mathbf{a}_{p_1}+\cdots+\mathbf{a}_{p_l})^2 F_{p_1}\cdots F_{p_l}}{\displaystyle 2d\sum_{l=0}^{\infty}(-)^l \sum_{p_1\neq\cdots\neq p_l} (T_{p_1}+\cdots+T_{p_l}) F_{p_1}\cdots F_{p_l}}\ , \tag{28}$$

with $F_p = \prod_{i=1}^{u} |\Lambda_{ip}|^{-1}$.

We remark that the existence and analyticity of Fredholm determinants such as (27) has recently been studied for hyperbolic diffeomorphism of finite smoothness on the basis of a new theory of distributions associated with the unstable and stable leafs of the diffeomorphism [64].

C. The nonequilibrium steady states

At the phenomenological level, a nonequilibrium steady state can be obtained from a hydrodynamic mode in the limit where the wavelength increases indefinitely together with the amplitude because $\lim_{L\to\infty}(L/2\pi)\sin(2\pi\mathbf{g}\cdot\mathbf{r}/L) = \mathbf{g}\cdot\mathbf{r}$, where $\mathbf{g} = \nabla c$ is a gradient of concentration c. Accordingly a nonequilibrium steady state can be obtained by the following limit [19]:

$$\Psi_{\mathrm{nss}} \equiv -\,i\,\mathbf{g}\cdot\partial_{\mathbf{k}}\Psi_{\mathbf{k}}\Big|_{\mathbf{k}=0}\ . \tag{29}$$

Since the first derivatives of the hydrodynamic mode is given by Eq. (23) and since the jump vector is related to the time integral of the particle velocity by Eq. (14), it is possible to obtain the following expression for the phase-space density of a nonequilibrium steady state with a gradient of concentration $\mathbf{g} = \nabla c$:

$$\Psi_{\mathrm{nns}}(\mathbf{X}) = \mathbf{g}\cdot\left[\mathbf{r}(\mathbf{X}) + \int_0^\infty \mathbf{v}(\mathbf{\Phi}^t\mathbf{X})\ dt\right]\ , \tag{30}$$

where $\mathbf{r}$ and $\mathbf{v}$ are respectively the position and the velocity of the tracer particle which diffuses [25].

This expression can also be derived from the phase-space probability density of a nonequilibrium steady state of an open system between two reservoirs of particles at different densities [33]. These reservoirs have the effect to impose flux boundary conditions to the diffusive system. As we said in Sec. 2, the discontinuities of this probability density occur on the unstable manifolds of the fractal repeller of the escape-rate formalism [33]. In the limit where the reservoirs are separated by an arbitrarily large distance $L \to \infty$ while keeping constant the gradient $\mathbf{g}$, the expression (30) is obtained for the phase-space density with respect to the density at a point in the middle of the system. The expression (30) is known as the Lebowitz-McLennan-Zubarev nonequilibrium steady state [65–68]. This density is an invariant of motion in the sense that $\{H, \Psi_{\rm nss}\} = 0$ where $\{H, \cdot\}$ is the Poisson bracket with the Hamiltonian.

An essential property of the nonequilibrium steady state is its singular character which has appeared in the limit $L \to \infty$. Indeed, Eq. (30) defines the density of a measure which is singular with respect to the Lebesgue measure. This singular character is furthermore of fractal type because the dynamics is chaotic. The fractal-like singular character is evidenced by considering the cumulative function associated with the density (30). For the so-called multibaker model, this cumulative function is given in terms of the continuous but nondifferentiable Takagi function which is self-similar [19]. This self-similarity is a reflect of the self-similarity of the underlying fractal repeller in a finite but large open system.

A consequence of Eq. (30) is Fick's law:

$$\mathbf{j} = -D \, \nabla c \, , \tag{31}$$

obtained by computing the mean flux $\mathbf{j}$ of particles for the nonequilibrium steady state (30) [19, 25].

D. Entropy production

The fractal-like singular character of the nonequilibrium steady state in the limit of a large system has a fundamental consequence on the question of entropy production. Indeed, the usual argument leading to the constancy of Gibbs' entropy becomes questionable because this argument assumes the existence of a density *function* for the probability density of the system. Since the density function may no longer exist in the limit $L \to \infty$, we may expect a very different behavior for the time variation of the entropy [34].

As shown very recently [69], a similar problem is expected for the time-dependent relaxation toward equilibrium in a finite system. In the long-time limit $t \to \infty$, the relaxation is described in terms of the hydrodynamic modes which are singular with respect to the Lebesgue measure so that the constancy of the entropy is here again in question.

A detailed analysis of the entropy production in an elementary model of diffusion known as the multibaker map has shown that the fractal-like singular character of the nonequilibrium steady state [34] – and equivalently of the hydrodynamic modes [69] – explains why the entropy production is positive and has the following form given by nonequilibrium thermodynamics [70]

$$\frac{d_i S}{dt} = \int D \, \frac{(\nabla c)^2}{c} \, d\mathbf{r} \; + \; \cdots \tag{32}$$

where the dots denote possible corrections of higher orders in the gradient. The calculation leading to this result simply assumes a coarse-grained entropy based on a partition of phase space into arbitrarily small cells [34]. Taking an arbitrarily fine partition allows us to get rid of the arbitrariness of the partition, in analogy with the procedure used to define the Kolmogorov-Sinai entropy per unit time.

For nonequilibrium steady states, the positive entropy production (32) is obtained in the limit where an arbitrarily large system is considered before an arbitrarily fine partition [34]. Indeed, for a finite system in a nonequilibrium steady state, there is always a small scale in phase space below which the invariant measure is continuous with respect to the Lebesgue measure, so that the entropy production vanishes for fine enough partitions. The remarkable fact is that this critical scale decreases exponentially rapidly in chaotic systems so that the behavior predicted by nonequilibrium thermodynamics will predominate even in relatively small systems. This result is especially interesting because it justifies the use of nonequilibrium thermodynamics already in small parts of a biological system.

For the time-dependent relaxation toward the equilibrium in chaotic systems, the positive entropy production (32) will be obtained in the long-time limit $t \to \infty$ for any partition into arbitrarily small cells, even if the system is finite [69]. This remarkable result has its origin in the fact that the hydrodynamic modes controlling the relaxation are always singular with respect to the Lebesgue measure even in finite chaotic systems. In this regard, we may conclude that the second law of thermodynamics is a consequence of the fractal-like singular character of the hydrodynamic modes which describe the relaxation toward the equilibrium. This conclusion is natural in view of the fact that this fractal-like singular character is a direct consequence of the phase-space mixing induced by the dynamical chaos.

V. CONCLUSIONS

In this short overview, we have summarized recent results about transport by diffusion in spatially periodic chaotic systems. These results extend to reaction-diffusion systems as recently shown elsewhere [71–73].

In this context, we have shown that all the relevant properties of transport (as well as of chemical reaction) are the consequences of a Frobenius-Perron operator such as Eq. (17). The generalized eigenvalue problem based on this Frobenius-Perron operator defines the hydrodynamic modes as the slowly damped long-wavelength eigenstates corresponding to the smallest Pollicott-Ruelle resonance $s_{\mathbf{k}}$. These modes describe the relaxation toward equilibrium of quasiperiodic inhomogeneities in the phase-space probability density of a statistical ensemble of trajectories. The inhomogeneities are quasiperiodic in the sense that the wavelength $L = 2\pi/\|\mathbf{k}\|$ of the hydrodynamic mode adds an extra periodicity to the intrinsic one of the lattice $\mathcal{L}$.

In systems with a gas of independent particles such as the Lorentz gas, the Frobenius-Perron operator (17) directly describes the relaxation toward equilibrium of quasiperiodic inhomogeneities in the position-velocity density of particles composing the gas. Therefore, our results show explicitly how the relaxation to equilibrium can occur in a N-particle system in the limit $N \to \infty$. Indeed, in this limit, the particle density $\rho(\mathbf{X})$ – or its Fourier component $f(\mathbf{x})$ – becomes a smooth function which evolves according to the Frobenius-Perron operator (16) – or (17) – so that the time evolution of the gas has the nonequilibrium properties described in Sec. 4.

A comment is here in order that the Pollicott-Ruelle resonances $s_{\mathbf{k}}$ are defined for the semigroup of the forward time evolution for $t \to +\infty$. By time reversibility, there exists another semigroup for $t \to -\infty$ with time-reversed properties. The hydrodynamic modes of the forward semigroup are singular in the stable directions of the phase space but they are smooth in the unstable directions because the dynamics is expanding in the unstable directions. In contrast, the hydrodynamic modes of the backward semigroup are singular in the unstable directions and smooth in the stable directions. This inequivalence constitutes a spontaneous breaking of the time-reversal symmetry which is due to the Lyapunov dynamical instability. We notice that a dynamical instability without chaos (like in the two-disk scatterer or in the inverted harmonic potential [11]) is already enough to generate this spontaneous time-reversal symmetry breaking. However, chaos is required to give a fractal character to the singular conditionally invariant measures of the hydrodynamic modes, which is essential to obtain the transport properties as well as for the entropy production (32).

We remark that the hydrodynamic modes exist only in the asymptotic expansions for $t \to \pm\infty$ of the time averages $\langle A \rangle_t$ of the physical observables $A(\mathbf{X})$. The relaxation toward equilibrium can be described in terms of exponentially decaying modes thanks to these time asymptotic expansions. Because of the dynamical instability, the forward asymptotic expansion turns out to be inequivalent to the backward asymptotic expansion. The forward asymptotic expansion for $t \to +\infty$ is obtained by the analytic continuation to complex frequencies $\exp(st) = \exp(i\omega t)$ with $\text{Im}\ \omega > 0$, while the backward expansion for $t \to -\infty$ is given by the analytic continuation to complex frequencies with $\text{Im}\ \omega < 0$. Supposing that the initial time of the experiment is taken at $t = 0$, it is a known result that the forward (resp. backward) asymptotic expansion converges only for $t > 0$ (resp. $t < 0$) [11]. Therefore, the origin $t = 0$ constitutes a horizon if we want to use the backward semigroup for future predictions on the time evolution of a statistical ensemble (or of a gas of independent particles). Somehow, this horizon is to the statistical ensemble of trajectories (or to the gas) what the Lyapunov horizon (1) is to a single trajectory. We observe that the accumulation of infinitely many trajectories in the statistical ensemble (or infinitely many particles in the gas) has created a horizon which is much stronger than the movable Lyapunov horizon (1). The semigroup horizon at $t = 0$ restricts the use of the exponentially decaying hydrodynamic modes to the future relaxation for $t > 0$. Therefore, this horizon constitutes a fundamental limit on the use of nonequilibrium thermodynamics in the description of many-particle systems. In this sense, the semigroup horizon justifies the irreversible character of nonequilibrium thermodynamics.

Acknowledgements

The author thanks Professor G. Nicolis for his support and encouragement in this research, as well as Professors J. R. Dorfman and S. Tasaki for many fruitful and inspiring discussions over the years. The "Fonds National de la Recherche Scientifique" is gratefully acknowledged for financial support. This work has also been financially supported by the InterUniversity Attraction Pole Program of the Belgian Federal Office of Scientific, Technical and Cultural Affairs.

[1] I. Prigogine, *Nonequilibrium Statistical Mechanics* (Wiley, New York, 1962).

[2] R. Balescu, *Equilibrium and Nonequilibrium Statistical Mechanics* (Wiley, New York, 1975).
[3] P. Résibois and M. De Leener, *Classical Kinetic Theory of Fluids* (Wiley, New York, 1977).
[4] J.-P. Boon and S. Yip, *Molecular Hydrodynamics* (Dover, New York, 1980).
[5] N. N. Krylov, Nature **153** (1944) 709; N. N. Krylov, *Works on the Foundations of Statistical Mechanics* (Princeton University Press, 1979); Ya. G. Sinai, *ibid.* p. 239.
[6] P. Gaspard and G. Nicolis, Physicalia Magazine (J. Belg. Phys. Soc.) **7** (1985) 151.
[7] R. Livi, A. Politi, and S. Ruffo, J. Phys. A: Math. Gen. **19** (1986) 2033.
[8] H. A. Posch and W. G. Hoover, Phys. Rev. A **38** (1988) 473.
[9] H. van Beijeren, J. R. Dorfman, H. A. Posch, and Ch. Dellago, Phys. Rev. E **56** (1997) 5272.
[10] N. I. Chernov, J. Stat. Phys. **88** (1997) 1.
[11] P. Gaspard, *Chaos, Scattering and Statistical Mechanics* (Cambridge University Press, Cambridge UK, 1998).
[12] J. R. Dorfman, *An Introduction to Chaos in Nonequilibrium Statistical Mechanics* (Cambridge University Press, Cambridge UK, 1999).
[13] L. A. Bunimovich and Ya. G. Sinai, Commun. Math. Phys. **78** (1980) 247, 479.
[14] N. I. Chernov, J. Stat. Phys. **74** (1994) 11.
[15] A. Knauf, Commun. Math. Phys. **110** (1987) 89; A. Knauf, Ann. Phys. (N. Y.) **191** (1989) 205.
[16] P. Gaspard, J. Stat. Phys. **68** (1992) 673.
[17] P. Gaspard, Chaos **3** (1993) 427.
[18] P. Gaspard, in: Y. Aizawa, S. Saito, and K. Shiraiwa, *Dynamical Systems and Chaos*, vol. 2 (World Scientific, Singapore, 1995) pp. 55-68.
[19] S. Tasaki and P. Gaspard, J. Stat. Phys. **81** (1995) 935.
[20] A. Baranyai, D. J. Evans, and E. G. D. Cohen, J. Stat. Phys. **70** (1993) 1085.
[21] N. I. Chernov, G. L. Eyink, J. L. Lebowitz, and Ya. G. Sinai, Phys. Rev. Lett. **70** (1993) 2209.
[22] T. Tél and J. Vollmer, *Entropy Balance, Multibaker Maps, and the Dynamics of the Lorentz Gas*, in: D. Szasz, Editor, *Hard Ball Systems and Lorentz Gas*, Encycl. Math. Sci. (Springer, Berlin, 2000).
[23] S. Tasaki and P. Gaspard, Theoretical Chemistry Accounts **102** (1999) 385.
[24] S. Tasaki and P. Gaspard, J. Stat. Phys. **101** (2000) 125.
[25] P. Gaspard, Phys. Rev. E **53** (1996) 4379.
[26] G. Nicolis, *Introduction to nonlinear science* (Cambridge University Press, Cambridge UK, 1995).
[27] J.-P. Eckmann and D. Ruelle, Rev. Mod. Phys. **57** (1985) 617.
[28] P. Gaspard, M. E. Briggs, M. K. Francis, J. V. Sengers, R. W. Gammon, J. R. Dorfman, and R. V. Calabrese, Nature **394** (1998) 865.
[29] P. Gaspard and S. A. Rice, J. Chem. Phys. **90** (1989) 2225; **91** (1989) E3279.
[30] P. Gaspard and G. Nicolis, Phys. Rev. Lett. **65** (1990) 1693.
[31] P. Gaspard and F. Baras, Phys. Rev. E **51** (1995) 5332.
[32] J. R. Dorfman and P. Gaspard, Phys. Rev. E **51** (1995) 28.
[33] P. Gaspard, Physica A **240** (1997) 54.
[34] P. Gaspard, J. Stat. Phys. **88** (1997) 1215.
[35] P. D. Lax and R. S. Phillips, *Scattering Theory* (Academic Press, New York, 1967).
[36] M. Pollicott, Invent. Math. **81** (1985) 413.
[37] M. Pollicott, Invent. Math. **85** (1986) 147.
[38] D. Ruelle, Phys. Rev. Lett. **56** (1986) 405.
[39] D. Ruelle, J. Stat. Phys. **44** (1986) 281.
[40] D. Ruelle, J. Diff. Geom. **25** (1987) 99, 117.
[41] D. Ruelle, Commun. Math. Phys. **125** (1989) 239.
[42] P. Gaspard and D. Alonso Ramirez, Phys. Rev. A **45** (1992) 8383.
[43] P. Gaspard, G. Nicolis, A. Provata, and S. Tasaki, Phys. Rev. E **51** (1995) 74.
[44] A. V. Andreev, O. Agam, B. D. Simons, and B. L. Altshuler, Phys. Rev. Lett. **76** (1996) 3947.
[45] O. Agam, Phys. Rev. E **61** (2000) 1285; G. Blum and O. Agam, Phys. Rev. E **62** (2000) 1977.
[46] M. Khodas, S. Fishman, and O. Agam, Phys. Rev. E **62** (2000) 4769.
[47] T. Kottos and U. Smilansky, Phys. Rev. Lett. **85** (2000) 968.
[48] J. Weber, F. Haake, and P. Seba, Phys. Rev. Lett. **85** (2000) 3620.
[49] F. Haake, *Quantum Signatures of Chaos*, 2nd Edition (Springer-Verlag Telos, Berlin, 2001).
[50] K. Pance, W. Lu, and S. Sridhar, Phys. Rev. Lett. **85** (2000) 2737.
[51] W. Lu, L. Viola, K. Pance, M. Rose, and S. Sridhar, Phys. Rev. E **61** (2000) 3652.
[52] S. Tasaki, I. Antoniou, and Z. Suchanecki, Phys. Lett. A **179** (1993) 97.
[53] H. H. Hasegawa and D. J. Driebe, *Phys. Rev. E* **50** (1994) 1781.
[54] D. J. Driebe, *Fully Chaotic Maps and Broken Time Symmetry* (Kluwer, Dordrecht, 1999).
[55] T. Gilbert, J. R. Dorfman, and P. Gaspard, Nonlinearity **14**, 339 (2001).
[56] P. Gaspard, I. Claus, T. Gilbert, and J. R. Dorfman, Phys. Rev. Lett. **86**, 1506 (2001).
[57] N. I. Chernov and C. P. Dettmann, Physica A **279** (2000) 37.
[58] C. P. Dettmann, *The Burnett expansion of the periodic Lorentz gas* (preprint `nlin.CD/0003038`, server `xxx.lanl.gov`).
[59] P. Cvitanović, Phys. Rev. Lett. **61** (1988) 2729.
[60] P. Cvitanović, R. Artuso, R. Mainieri, and G. Vattay, `http://www.nbi.dk/ChaosBook/`

[61] R. Artuso, Phys. Lett. A **160** (1991) 528.
[62] P. Cvitanović, P. Gaspard, and T. Schreiber, Chaos **2** (1992) 85.
[63] P. Cvitanović, J.-P. Eckmann, and P. Gaspard, Chaos, Solitons & Fractals **6** (1995) 113.
[64] A. Yu. Kitaev, Nonlinearity **12** (1999) 141.
[65] J. L. Lebowitz, Phys. Rev. **114** (1959) 1192.
[66] J. A. MacLennan Jr., Phys. Rev. **115** (1959) 1405.
[67] D. N. Zubarev, Sov. Phys. Dokl. **6** (1962) 776.
[68] D. N. Zubarev, *Nonequilibrium Statistical Thermodynamics* (Consultants, New York, 1974).
[69] T. Gilbert, J. R. Dorfman, and P. Gaspard, Phys. Rev. Lett. **85** (2000) 1606.
[70] I. Prigogine, *Introduction to the Thermodynamics of Irreversible Processes* (Wiley, New York, 1961).
[71] P. Gaspard and R. Klages, Chaos **8** (1998) 409.
[72] P. Gaspard, Physica A **263** (1999) 315.
[73] I. Claus and P. Gaspard, J. Stat. Phys. **101** (2000) 161.