From dynamical systems theory to nonequilibrium thermodynamics

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An overview is given of recent advances on diffusion and nonequilibrium thermodynamics in the perspective given by the pioneering work of Henri Poincaré on dynamical systems theory. The hydrodynamic modes of diffusion are explicitly constructed from the underlying deterministic dynamics in the multibaker, the hard-disk and Yukawa-potential Lorentz gases, as well as in a geodesic flow on a noncompact manifold of constant negative curvature built out of the Poincaré disk. These modes are represented by singular distributions with fractal cumulative functions.

I. INTRODUCTION

The purpose of the present paper is to give an overview of the contributions of Henri Poincaré to dynamical systems theory and its modern developments in the theory of chaos and our current understanding of the dynamical bases of nonequilibrium thermodynamics.

Poincaré is the founding father of modern dynamical systems theory by his monumental work on celestial mechanics. The fundamental concepts he has introduced still play a central role in this field and beyond. By inventing the concept of homoclinic orbit and by describing for the first time a homoclinic tangle, Poincaré discovered what is today referred to as dynamical chaos. This is a particular state of motion in which most of the trajectories are random although they are solutions of a deterministic dynamical systems defined in terms of regular differential equations or maps. Chaotic motion has been discovered in Hamiltonian, conservative, and dissipative dynamical systems with a few or many degrees of freedom. Many disciplines are concerned with dynamical chaos from mathematics to natural sciences such as physics, chemistry, meteorology and geophysics, or biology where many examples of chaotic systems have been discovered and studied.

In the present paper, the focus will be on dynamical systems of interest for the transport properties and, in particular, the property of diffusion, which is of interest for nonequilibrium statistical mechanics. Diffusion is a process of transport of particles across a spatially extended system. If the motion is chaotic, the particle may perform a random walk due to its successive collisions with scatterers. The process of diffusion is described in nonequilibrium thermodynamics as an irreversible process responsible for entropy production. For this reason, deterministic dynamical systems with diffusion constitute interesting models to investigate mechanisms leading to irreversible behavior as described by nonequilibrium thermodynamics. This program has been successfully carried out for simple models, leading to a very precise understanding of entropy production in the case of diffusion [1–6]. It has been discovered that it is possible to construct so-called hydrodynamic modes of diffusion from the chaotic dynamics. These modes are defined as generalized eigenstates of the Liouvillian or Frobenius-Perron operators ruling the time evolution of statistical ensembles of trajectories. The associated eigenvalues, also called the Pollicott-Ruelle resonances, give the damping rates of the modes and they control the relaxation toward the state of thermodynamics equilibrium. These generalized eigenstates are not given by regular functions contrary what might be expected: they are given by singular mathematical distributions with fractal properties. The singular character of these eigenstates has been shown to be responsible for the entropy irreversibly produced by diffusion during the relaxation toward equilibrium, as expected from nonequilibrium thermodynamics. The irreversible character is understood by the fact that the eigenstates break the time-reversal symmetry. Indeed, the eigenstates are smooth along the unstable manifolds of the chaotic dynamics but singular along the stable manifolds. Therefore, they are distinct from their image under time reversal.

In the present paper, these eigenstates representing the hydrodynamic modes of diffusion are presented for different systems: the multibaker map which is a simplified model of diffusion, the hard-disk and Yukawa-potential Lorentz gases, and finally for a geodesic flow on a negative curvature space. This last example is constructed in the Poincaré disk of nonEuclidean geometry. All these systems have in common to be spatially periodic so that their Liouvillian dynamics can be analyzed by spatial Fourier transforms, leading to the explicit construction of the Liouvillian eigenstates and associated eigenvalues. These results provide one of the most advanced understanding of the dynamical bases of kinetic theory and nonequilibrium thermodynamics.

The plan of the paper is the following. In Sec. II, an overview is given of Poincaré’s contributions to dynamical
systems theory. Remarkably, Poincaré has also contributions to gas kinetic theory which are reviewed in Sec. III. In Sec. IV, it is shown that dynamical chaos is very common in systems of statistical mechanics. The aim of Sec. V is to explain how time-reversal symmetry can be broken in a theory based on an equation having the invariance. The construction of the hydrodynamic modes of diffusion is carried out in Sec. VI where they are shown to have fractal properties. This construction is applied to the multibaker map and the Lorentz gases in Sec. VII. In Sec. VIII, diffusion is presented for a geodesic flow on a negative curvature space. Finally, the implications for entropy production and nonequilibrium thermodynamics are discussed in Sec. IX and conclusions are drawn in Sec. X.

II. POINCARÉ AND DYNAMICAL SYSTEMS THEORY

Henri Poincaré is considered as the founding father of dynamical systems theory because he has discovered many basic results of immediate use for the qualitative and quantitative analyses of dynamical systems. The advent of modern computers has allowed us to explicitly and numerically construct the phase-space objects discovered by Poincaré. Poincaré's advances into dynamical systems theory are due to his efforts to understand the stability of the Solar System and, in particular, the problem of three bodies in gravitational interaction. His memoir for the King Oscar Prize in 1888 has led to the publications in 1892, 1893 and 1899 of his famous three-volume work entitled Les méthodes nouvelles de la mécanique céleste (The novel methods of celestial mechanics). Today, this work is still the source of inspiration for active research in celestial mechanics and dynamical systems theory. Some of the most recent spatial missions are using complicated orbits meandering through phase-space chaotic zones in order to minimize fuel. Some of the main contributions of Poincaré to dynamical systems theory are the following results and concepts:

- the Poincaré-Bendixson theorem which proves the absence of chaos in two-dimensional flows;
- the Poincaré surface of section which is a basic tool for the study of phase-space geometry;
- the stable $W_s(\gamma)$ and unstable $W_u(\gamma)$ invariant manifolds of an orbit $\gamma$;
- the homoclinic orbit which is defined by Poincaré as the intersection $W_s(\gamma) \cap W_u(\gamma)$ between the stable and unstable manifolds of an orbit $\gamma$, as well as the heteroclinic orbit defined as the intersection $W_u(\gamma) \cap W_u(\gamma')$ of the stable manifold of an orbit $\gamma$ with the unstable manifold of another orbit $\gamma'$;
- the homoclinic tangle which is the phase-space figure formed by the intersecting stable and unstable manifolds of a set of orbits and which is at the origin of chaotic behavior in three-dimensional flows. Poincaré gave a description of the homoclinic tangle in his book but only modern work has provided concrete visualization of homoclinic tangles.

These concepts were developed later, notably, by Birkhoff and Shil'nikov who proved the existence of periodic and nonperiodic orbits in the vicinity of a homoclinic orbit. This led to the notion of symbolic dynamics in which sequences of symbols can be associated with the trajectories of the system. In this way, it became possible to list and enumerate the trajectories. If the sequences are freely built on the basis of an alphabet with several symbols, the periodic orbits proliferate exponentially as their period increases. The rate of exponential proliferation is the so-called topological entropy per unit time, which provides a quantitative topological characterization of chaotic behavior. These properties concern both conservative and dissipative dynamical systems.

III. POINCARÉ AND GAS KINETIC THEORY

Poincaré published few papers on gas kinetic theory. They reflect the extraordinary dynamism of Poincaré to follow the scientific developments of his time but, contrary to other fields where Poincaré could anticipate, his papers on gas kinetic theory appear as comments on previous contributions by others. In the years 1892-1894, Poincaré wrote a few critical comments about Maxwell’s approach, which attests that a definitive mathematical formulation of the experimental facts was still lacking in those years [7].

Later, Poincaré published in 1906 a paper entitled Réflexions sur la théorie cinétique des gaz (Thoughts on gas kinetic theory) [8], which cites Gibbs’ contribution of 1902 [9]. This paper contains a version of what is today called Liouville’s equation. For the time evolution of the phase-space probability density $P(\{x_i\}, t)$, Poincaré uses the continuity equation

$$\frac{\partial P}{\partial t} + \sum_i \frac{\partial (P X_i)}{\partial x_i} = 0 \tag{1}$$
corresponding to a dynamical system defined by the ordinary differential equations

$$\frac{dx_i}{dt} = X_i$$

(2)

For a system obeying Liouville's theorem

$$\sum_i \frac{\partial X_i}{\partial x_i} = 0$$

(3)

he also gives the form

$$\frac{\partial P}{\partial t} + \sum_i X_i \frac{\partial P}{\partial x_i} = 0$$

(4)

which Gibbs wrote in 1902 as

$$\frac{\partial P}{\partial t} = - \sum_k \left( \frac{\partial P}{\partial p_k} \dot{p}_k + \frac{\partial P}{\partial q_k} \dot{q}_k \right)$$

(5)

with \( \{x_i\} = \{(p_k, q_k)\} \) [9]. A consensus on the basic equation of statistical mechanics is thus clearly emerging during these years. Liouville’s equation can also be written as

$$\frac{\partial P}{\partial t} = \{H, P\} \equiv \hat{L}P$$

(6)

in terms of the Poisson bracket \( \{., .\} \) of the Hamiltonian \( H \) with the probability density \( P \), which defines the Liouvillian operator \( \hat{L} \).

Besides, Poincaré’s 1906 paper mainly contains a discussion of the behavior of the coarse-grained and fine-grained entropies in an ideal gas in the absence or presence of an external perturbation. For this model, Poincaré discusses in particular about the constancy of the fine-grained entropy in the absence of external perturbation and the increase in time of the coarse-grained entropy. The conclusions of the paper expresses Poincaré’s appreciation that, with such results, the last difficulties of gas kinetic theory finally disappear [8].

Poincaré also published a treatise on thermodynamics which contains at the end a discussion of his viewpoint about the deduction of the principles of thermodynamics from those of mechanics [10].

It should be added that Poincaré published his famous recurrence theorem in 1890 in the context of the three-body problem and it was Zermelo who, in 1896, used this theorem as an objection to Boltzmann’s \( H \)-theorem, a debate to which Poincaré did not directly participate.

IV. CHAOTIC BEHAVIOR IN MOLECULAR DYNAMICS

The merging of dynamical systems theory and chaos theory with modern statistical mechanics occurred much later. During the twenties and the thirties, ergodic theory underwent important developments with contributions from Birkhoff, Hedlund, Hopf, Koopman, von Neumann, Seidel, and others. On the one hand, these developments contributed to define rigorously the notions of ergodicity and of mixing and, on the other hand, they showed that these statistical properties manifest themselves in hyperbolic dynamical systems such as geodesic motions on closed surfaces of constant negative curvature.

In the forties, Krylov proposed such hyperbolic motions as models for particles interacting in a gas. At Los Alamos, von Neumann and Ulam pointed out that chaotic maps such as the logistic map \( x_{n+1} = 4x_n(1-x_n) \) can be used as random generators for Monte Carlo calculations. In the seventies, Sinai proved the ergodicity and mixing of some hard-ball models of gases (see Fig. 1). In this way, he also proved that hard-ball gases are chaotic and have the property of sensitivity to initial conditions. The concept of Lyapunov exponent was defined as

$$\lambda_i = \lim_{t \to \infty} \frac{1}{t} \ln \| \delta x^{(i)}(t) \| \| \delta x^{(i)}(0) \|$$

(7)

where \( \delta x^{(i)}(t) \) denotes a perturbation on the phase-space trajectory \( x(t) \) and pointing in one of all the possible phase-space directions. In 1974, Erpenbeck carried out the simulation of molecular dynamics for systems of interacting
particles on the Los Alamos computers and reported their high sensitivity to initial conditions. In the eighties, the first estimations of the value of these exponents in typical systems of statistical mechanics were published. Following Krylov’s reasoning, the largest Lyapunov exponent was shown to take the very high value

$$\lambda \sim \frac{1}{\tau} \ln \frac{\ell}{d} \sim 10^{10} \text{s}^{-1}$$  (8)

for air at room temperature and pressure where $d$ is the diameter of the particles, $\tau$ the intercollisional time, and $\ell$ the mean free path (cf. Fig. 2).

FIG. 1: Schematic representation of a dynamical system of hard spheres in elastic collisions and moving on a torus $T^3$.

FIG. 2: Amplification of some perturbation on the velocity angle $\delta \phi_n$ for a particle undergoing two successive collisions separated on average with a distance of the order of the mean free path $\ell$. The particles have a diameter $d$. After the second collision, the perturbation on the velocity angle is amplified by a factor given by the ratio of the mean free path over the particle diameter: $\delta \phi_{n+1} \sim (\ell/d) \delta \phi_n$. Since the time between the two collisions is of the order of the intercollisional time $\tau$, the maximum Lyapunov exponent is estimated to be $\lambda \sim (1/\tau) \ln (\ell/d)$.

The systematic numerical calculation of the spectrum of Lyapunov exponents was undertaken in the eighties and, in the nineties, theoretical evaluations were obtained by van Beijeren, Dorfman, and others. Today, it is a routine calculation to obtain the spectrum of Lyapunov exponents for a system of statistical mechanics (see Fig. 3). The sum of positive Lyapunov exponents gives the so-called Kolmogorov-Sinai entropy per unit time according to Pesin’s formula

$$h_{\text{KS}} = \sum_{\lambda_i > 0} \lambda_i$$  (9)

The Kolmogorov-Sinai entropy per unit time characterizes the dynamical randomness displayed by the trajectories of the total system of particles. It is the analogue of the standard entropy per unit volume obtained by replacing space by time. In this regard, the Kolmogorov-Sinai entropy per unit time is a measure of time disorder in the dynamics of the system. The Kolmogorov-Sinai entropy is defined for the ergodic invariant probability measure of the thermodynamic equilibrium. It gives a lower bound on the topological entropy per unit time

$$h_{\text{KS}} \leq h_{\text{top}}$$  (10)

which is the rate of proliferation of unstable periodic orbits in the system. Each of these periodic orbits has a stable manifold and an unstable manifold extending in the phase space and intersecting with each other. The positive
topological entropy per unit time inferred from Eqs. (9) and (10) implies the existence of an important Poincaré homoclinic tangle in the high-dimensional phase space of the systems of interacting particles.

\[ \text{FIG. 3: Spectrum of Lyapunov exponents of a dynamical system of 33 hard spheres of unit diameter and mass at unit temperature and density 0.001. The Lyapunov exponents obey the pairing rule that the Lyapunov exponents come in pairs } \{\lambda_i, -\lambda_i\}. \text{ Eight Lyapunov exponents vanish because the system has four conserved quantities, namely, energy and the three components of momentum and because of the pairing rule. The total number of Lyapunov exponents is equal to } 6 \times 33 = 198. \]

V. SPONTANEOUS BREAKING OF TIME-REVERSAL SYMMETRY AND POLLICOTT-RUELLE RESONANCES

Hamilton’s equations of motion are symmetric under time reversal \( \Theta(p_k, q_k) = (-p_k, q_k) \). However, it is known that the solution of an equation may have a lower symmetry than the equation itself. Accordingly, some invariant subsets of phase space do not need to be time-reversal symmetric. For instance, Poincaré’s invariant stable and unstable manifolds of an orbit \( \gamma \) or of a set of orbits are not symmetric under time reversal:

\[ \Theta W_s = W_u \neq W_s \]

This suggests that irreversible behavior may result from weighting differently the stable \( W_s \) and unstable \( W_u \) manifolds with a measure. This idea is at the basis of the concept of Pollicott-Ruelle resonance which has been rigorously defined for Axiom-A systems [11, 12]. An Axiom-A system \( \Phi^t \) satisfies the properties that: (1) The nonwandering set \( \Omega(\Phi^t) \) is hyperbolic; (2) The periodic orbits of \( \Phi^t \) are dense in \( \Omega(\Phi^t) \). These systems are known to be strongly chaotic and mixing so that the mean values of the observables typically decay in time. The Pollicott-Ruelle resonances provide the decay rates of the time evolution. These resonances are the classical analogues of the quantum scattering resonances since they are the poles of the resolvent of the Liouvillian operator [13]

\[ \frac{1}{s - \hat{L}} \]

These poles can be obtained by analytic continuation toward complex frequencies (see Fig. 4). Accordingly, the Pollicott-Ruelle resonances can be conceived as the generalized eigenvalues of Liouville’s equation. The associated decaying eigenstates are singular in the direction of \( W_s \) but smooth in \( W_u \). Since the so-extended Liouvillian operator is neither Hermitian nor anti-Hermitian, the right- and left-eigenstates are in general different:

\[ \hat{L} |\Psi_\alpha\rangle = s_\alpha |\Psi_\alpha\rangle \]

\[ \langle \Psi_\alpha | \hat{L} = s_\alpha \langle \Psi_\alpha | \]

with \( s_\alpha \) complex. Moreover, Jordan-blocks structures may arise from multiple eigenvalues.
FIG. 4: Pollicott-Ruelle resonances and antiresonances in the complex plane of the variable $s$.

A priori, the Liouvillian operator defines a group of time evolution defined over the whole time axis $-\infty < t < +\infty$. The mean values of an observable $A(x)$ evolves in time according to:

$$\langle A \rangle_t = \langle A | \exp(\hat{L} t) | P_0 \rangle = \int A(x) P_0(\Phi^{-t} x) \, dx$$

(15)

However, after having performed the analytic continuation toward either the lower or the upper half complex plane $s$, the group splits into two semigroups (see Fig. 5).

On the one hand, the forward semigroup valid for $0 < t < +\infty$ is given in terms of the resonances with $\text{Re } s_\alpha < 0$:

$$\langle A \rangle_t = \langle A | \exp(\hat{L} t) | P_0 \rangle \approx \sum_\alpha \langle A | \Psi_\alpha \rangle \exp(s_\alpha t) \langle \tilde{\Psi}_\alpha | P_0 \rangle + (\text{Jordan blocks})$$

(16)

This expression can be obtained as an asymptotic expansion around $t = +\infty$.

On the other hand, the backward semigroup valid for $-\infty < t < 0$ is given in terms of the antiresonances with
Re \( s_\alpha > 0 \):

\[
\langle A \rangle_t = \langle A \exp(\hat{L}t) | P_0 \rangle \approx \sum_\alpha \langle A | \Psi_\alpha \circ \Theta \rangle \exp(-s_\alpha t) \langle \tilde{\Psi}_\alpha \circ \Theta | P_0 \rangle + \text{(Jordan blocks)}
\]  

(17)

It is obtained as an asymptotic expansion around \( t = -\infty \). The time-reversal symmetry implies that a resonance \( s_\alpha \) corresponds to each antiresonance \( -s_\alpha \). However, the associated eigenstates \( \Psi_\alpha \) and \( \Psi_\alpha \circ \Theta \) differ because the former is smooth along the unstable manifolds though the latter is smooth along the stable ones. Hence, these eigenstates break the time-reversal symmetry. We are in the presence of a phenomenon of broken symmetry very similar to other such phenomena known in condensed-matter physics.

VI. DIFFUSION IN SPATIALLY EXTENDED SYSTEMS

We now consider dynamical systems which are spatially extended and which can sustain a transport process of diffusion. We moreover assume that the system is invariant under a discrete abelian subgroup of spatial translations \( \{a\} \). The invariance of the Liouvillian time evolution under this subgroup means that the Perron-Frobenius operator \( \hat{P} = \exp(\hat{L}t) \) commutes with the translation operators \( \hat{T}_a \):

\[
[\hat{P}, \hat{T}_a] = 0
\]  

(18)

Consequently, these operators admit common eigenstates:

\[
\begin{align*}
\hat{P}\psi_k &= \exp(s_k t)\psi_k \\
\hat{T}_a\psi_k &= \exp(ik \cdot a)\psi_k
\end{align*}
\]  

(19)

where \( k \) is called the wavenumber or Bloch parameter. The Pollicott-Ruelle resonances are now functions of the wavenumber. For strongly chaotic systems with two degrees of freedom, the resonances can be obtained as the roots of the dynamical zeta function:

\[
Z(s; k) = \prod_p \prod_{m=0}^{\infty} \left( 1 - \frac{\exp(-sT_p - ik \cdot a_p)}{|\Lambda_p|^{m+1}} \right)
\]  

(20)

which is given by a product over all the unstable periodic orbits \( p \) of instability eigenvalue \( \Lambda_p \) and prime period \( T_p \). The lattice vector \( a_p \) gives the spatial distance travelled by the particle along the prime period of the periodic orbit \( p \).

The leading Pollicott-Ruelle resonance which vanishes with the wavenumber defines the dispersion relation of diffusion:

\[
s_k = -Dk^2 + O(k^4)
\]  

(21)

The diffusion coefficient is given by the Green-Kubo formula:

\[
D = \int_0^{\infty} \langle v_x(0)v_x(t) \rangle \, dt
\]  

(22)

where \( v_x \) is the velocity of the diffusing particle. The associated eigenstate is the hydrodynamic mode of diffusion \( \Psi_k \).

A. Fractality of the hydrodynamic modes of diffusion

In strongly chaotic systems, the diffusive eigenstate \( \Psi_k \) is a Schwartz-type distribution which is smooth in the unstable direction \( W_u \) but singular in \( W_s \). This distribution cannot be depicted except by its cumulative function

\[
F_k(\theta) = \int_0^\theta \Psi_k(x'') \, d\theta'
\]  

(23)
defined by integrating over a curve \(x_0\) in the phase space (with \(0 \leq \theta < 2\pi\)). We notice that the eigenstate \(\Psi_k\) of the forward semigroup can be obtained by applying the time evolution operator over an arbitrary long time from an initial function which is spatially periodic of wavenumber \(k\) whereupon the cumulative function (23) is equivalently defined by

\[
F_k(\theta) = \lim_{t \to \infty} \frac{\int_0^\theta \, d\theta' \exp[i k \cdot (r_t - r_0)]}{\int_0^{2\pi} \, d\theta' \exp[i k \cdot (r_t - r_0)]}
\]

(24)

This function is normalized to take the unit value for \(\theta = 2\pi\). For vanishing wavenumber, the cumulative function is equal to \(F_k(\theta) = \theta/(2\pi)\), which is the cumulative function of the microcanonical uniform distribution in phase space. For nonvanishing wavenumbers, the cumulative function becomes complex.

Examples of such cumulative functions of the hydrodynamic modes of diffusion are depicted in the following for several dynamical systems of chaotic diffusion. These cumulative functions typically form fractal curves in the complex plane \((\text{Re } F_k, \text{Im } F_k)\). It has been proved that the Hausdorff dimension \(D_H\) of these fractal curves can be calculated by the formula:

\[
P(D_H) = D_H \text{ Re } s_k
\]

(25)
in terms of the Ruelle topological pressure:

\[
P(\beta) \equiv \lim_{t \to \infty} \frac{1}{t} \ln \langle |\Lambda_t|^{1-\beta} \rangle
\]

(26)

where \(\Lambda_t\) is the factor by which the phase-space volume are stretched along the unstable direction [5]. The positive Lyapunov exponent is given by

\[
\lambda = -P'(1) = \lim_{t \to \infty} \frac{1}{t} \langle \ln |\Lambda_t| \rangle
\]

(27)
The system is closed so that there is no escape of particles and \(P(1) = 0\). The Hausdorff dimension can be expanded in powers of the wavenumber as

\[
D_H(k) = 1 + \frac{D}{\lambda} k^2 + O(k^4)
\]

(28)

so that the diffusion coefficient can be obtained from the Hausdorff dimension and the Lyapunov exponent by the formula

\[
D = \lambda \lim_{k \to 0} \frac{D_H(k) - 1}{k^2}
\]

(29)

This formula has been verified for different dynamical systems sustaining deterministic diffusion [5].

VII. EXAMPLES OF DIFFUSIVE MODES

In the present section, we explicitly construct the hydrodynamic modes of diffusion for several spatially periodic dynamical systems. All the systems are of Hamiltonian character so that they are time-reversal symmetric and obey Liouville’s theorem. These systems have two degrees of freedom and are chaotic so that they have the Lyapunov spectrum \((\lambda, 0, 0, -\lambda)\) where \(\lambda\) is the unique positive Lyapunov exponent.

A. Multibaker model of diffusion

One of the simplest models of chaotic diffusion is the multibaker map which is a generalization of the well-known baker map into a spatially periodic system. The map is two-dimensional and rules the motion of a particle which can jump from square to square in a random walk. The equations of the map are given by [1, 2]

\[
\phi(l, x, y) = \begin{cases} 
(l - 1, 2x, \frac{y}{2}), & 0 \leq x \leq \frac{1}{2} \\
(l + 1, 2x - 1, \frac{y + 1}{2}), & \frac{1}{2} < x \leq 1
\end{cases}
\]

(30)
where \((x, y)\) are the coordinates of the particle inside a square, while \(l \in \mathbb{Z}\) is an integer specifying in which square the particle is currently located. This map acts as a baker map but, instead of mapping the two stretched halves into themselves, they are moved to the next-neighboring squares as shown in Fig. 6.

\[
\ldots \quad l \quad l \quad l \quad \ldots
\]

\[
\ldots \quad \text{gray} \quad \text{red} \quad \text{gray} \quad \ldots \quad \phi
\]

**FIG. 6**: Schematic representation of the multibaker map \(\phi\) acting on an infinite sequence of squares.

![Multibaker Map Diagram](image)

**FIG. 7**: The diffusive modes of the multibaker map represented by their cumulative function depicted in the complex plane \((\text{Re } F_k, \text{Im } F_k)\) versus the wavenumber \(k\).

The multibaker map preserves the vertical and horizontal directions, which correspond respectively to the stable and unstable directions. Accordingly, the diffusive modes of the forward semigroup are horizontally smooth but vertically singular. Both directions decouple and it is possible to write down iterative equations for the cumulative functions of the diffusive modes, which are known as de Rham functions [2]

\[
F_k(y) = \begin{cases} 
\alpha F_k(2y), & 0 \leq y \leq \frac{1}{2} \\
(1 - \alpha)F_k(2y - 1) + \alpha, & \frac{1}{2} < y \leq 1 
\end{cases}
\]  

(31)

with

\[
\alpha = \frac{\exp(ik)}{2 \cos k}
\]  

(32)

For each value of the wavenumber \(k\), the de Rham functions depict a nice fractal curve as seen in Fig. 7. The fractal dimension of these fractal curves can be calculated as follows. The dispersion relation of diffusion has the analytical expression

\[
s_k = \ln \cos k = -\frac{1}{2} k^2 + O(k^4)
\]  

(33)
so that the diffusion coefficient is equal to $D = \frac{1}{2}$. Since the dynamics is uniformly expanding by a factor 2 in the multibaker map, the Ruelle topological pressure has the form

$$P(\beta) = (1 - \beta) \ln 2$$

whereupon the Hausdorff dimension of the diffusive mode is given by

$$D_H = \frac{\ln 2}{\ln 2 \cos k}$$

according to Eq. (25) [4].

B. Periodic hard-disk Lorentz gas

The periodic hard-disk Lorentz gas is a two-dimensional billiard in which a point particle undergoes elastic collisions on hard disks which are fixed in the plane in the form of a spatially periodic lattice. Bunimovich and Sinai have proved that the motion is diffusive if the horizon seen by the particles is always finite [14]. This is the case for a hexagonal lattice under the condition that the disks are large enough to avoid the possibility of straight trajectories moving across the whole lattice without collision. The dynamics of this system is ruled by the free-particle Hamiltonian:

$$H = \frac{p^2}{2m}$$

supplemented by the rules of elastic collisions on the disks. Because of the defocusing character of the collisions on the disks, the motion is chaotic. Two trajectories from slightly different initial conditions are depicted in Fig. 8. The dynamics is very sensitive to the initial conditions because the trajectories separate after a few collisions as seen in Fig. 8b. On long times, the trajectories perform random walks on the lattice (see Fig. 8a).

The cumulative functions of the diffusive modes can be constructed by using Eq. (24). The trajectories start from the border of a disk with an initial position at an angle $\theta$ with respect to the horizontal and an initial velocity normal
The diffusive modes of the periodic hard-disk Lorentz gas represented by their cumulative function depicted in the complex plane $(\text{Re } F_k, \text{Im } F_k)$ versus the wavenumber $k$.

The results are depicted in Figs. 9 and 10 where we see the fractal character of these curves developing as the wavenumber $k$ increases. The Hausdorff dimension indeed satisfies Eq. (28) as shown elsewhere [5].

C. Periodic Yukawa-potential Lorentz gas

This other Lorentz gas is similar to the previous one except that the hard disks are replaced by Yukawa potentials centered here at the vertices of a square lattice. Knauf has proved that this system is chaotic and diffusive if the energy of the moving particles is large enough [15]. The Hamiltonian of this system is given by

$$H = \frac{p^2}{2m} - \sum_i \frac{\exp(-ar_i)}{r_i}$$

(37)
where $a$ is the inverse screening length. The sensitivity to initial conditions is illustrated in Fig. 11 which shows two trajectories starting from very close initial conditions. The particles undergo a random walk on long time scales.

FIG. 11: Two trajectories of the periodic Yukawa-potential Lorentz gas. They start from the same position but velocities which differ by one part in a million.

The cumulative functions of the diffusive modes can here also be constructed by using Eq. (24) with trajectories integrated with a numerical algorithm based on a rescaling of time at the singular collisions. The initial position is taken on a small circle around a scattering center at an angle $\theta$ with respect to the horizontal direction and the initial velocity is normal and pointing to the exterior of this circle. The results are depicted in Fig. 12 for two nonvanishing wavenumbers. The Hausdorff dimension of these fractal curves also satisfies Eq. (28) as shown elsewhere [5].

FIG. 12: The diffusive modes of the periodic Yukawa-potential Lorentz gas represented by their cumulative function depicted in the complex plane $(\text{Re } F_k, \text{Im } F_k)$ versus the wavenumber $k$. At the vanishing wavenumber $k = 0$, the curve reduces to a straight line corresponding to the invariant microcanonical equilibrium state.
VIII. DIFFUSION IN A GEODESIC FLOW ON A NEGATIVE CURVATURE SPACE

A further example of chaotic diffusion is provided by a geodesic flow on a compact manifold of negative curvature. This manifold is constructed in the non-Euclidean geometry of Bolyai and Lobatchevsky with the Riemannian metric:

\[ ds^2 = \text{d}r^2 + (\sinh \tau)^2 \text{d}\varphi^2 \]  

This space can be represented in the Poincaré disk (see Fig. 13). The Cartesian coordinates of the complex plane where the Poincaré disk is depicted are given by

\[ \zeta = \xi + i\eta = \exp(i\varphi) \tanh \frac{\tau}{2} \]  

In these coordinates, the metric (38) becomes

\[ ds^2 = 4 \frac{d\xi^2 + d\eta^2}{(1 - \xi^2 - \eta^2)^2} \]  

The line at infinity \((\tau = \infty)\) is the border of the disk \((\xi^2 + \eta^2 = 1)\). The geodesics of this space are the diameters of the disk or the arcs of circles which are perpendicular to the border of the disk.

![FIG. 13: Poincaré disk representing the Bolyai-Lobatchevsky space with two geodesics (see text).](image)

![FIG. 14: (a) Octogon \(D/G\) formed with the four boosts \(g_0, g_1, g_2,\) and \(g_3\) in the Poincaré disk. (b) The compact manifold \(D/G\) of genus two built by identification of the opposite sides of the octogon using the four boosts \(g_0, g_1, g_2,\) and \(g_3\) and their inverses.](image)

The Poincaré disk can be mapped onto the Poincaré half-plane by the following change of coordinates

\[ z = x + iy = i \frac{1 - \zeta}{1 + \zeta} \]  

in terms of which the Riemannian metric becomes:

\[ ds^2 = \frac{dx^2 + dy^2}{y^2} \]
The Poincaré half-plane is another representation of the non-Euclidean geometry of Bolyai and Lobatchevsky.

A compact manifold called the octogon can be built in the Poincaré disk $D$ by taking the quotient $D/G$ of the disk with a discrete group $G$ generated by the boosts $g_0$, $g_1$, $g_2$, and $g_3$ [16]. The first boost is defined by the matrix

$$g_0 = \begin{bmatrix} \cosh \frac{\tau}{2} & \sinh \frac{\tau}{2} \\ \sinh \frac{\tau}{2} & \cosh \frac{\tau}{2} \end{bmatrix} \in SU(1,1)$$

with $\cosh \frac{\tau}{2} = 1 + \sqrt{2}$. The boosts $g_1$, $g_2$, and $g_3$ are the rotations of $g_0$ by $\frac{\pi}{4}$, $\frac{\pi}{2}$, and $\frac{3\pi}{4}$, respectively. These four boosts and their inverses generate a non-Abelian discrete group $G$ of symmetries of the Poincaré disk. The basic relation of this group is

$$\left( g_0 g_1^{-1} g_2 g_3^{-1} \right) \left( g_0^{-1} g_1 g_2^{-1} g_3 \right) = 1$$

Each boost can be used to map onto each other two opposite circles perpendicular to the border of the disk as shown in Fig. 14a. The eight circles intersect to form a curvilinear octogon. The opposite sides of this octogon can be identified with the four boosts and their inverses in order to define a compact manifold of genus two depicted in Fig. 14b.

Since the manifold $D/G$ is compact it cannot sustain a transport process of diffusion. In order to allow for infinite trajectories, we periodically extend the octogon by using the Abelian subgroup of the discrete group $G$ generated by the boost $g_0$:

$$\{g_0^n\}_{n=-\infty}^{+\infty}$$

The images of the octogon generated by the boost $g_0$ and its iterates form a domain of the Poincaré disk extending up to infinity, as shown in Fig. 15. This domain forms the noncompact manifold of Fig. 16, which has a genus equal to infinity.

FIG. 15: Infinite domain of the Poincaré disk obtained by extending the octogon of Fig. 14 using the Abelian subgroup generated by the boost $g_0$. The picture on the left-hand side is a zoom on the left-hand side of the border of the Poincaré disk. The integers denote the number of boosts $g_0$ needed to reach the corresponding image of the octogon.

FIG. 16: The noncompact manifold of genus equal to infinity corresponding to the domain of Fig. 15.
FIG. 17: Random walk of a typical trajectory in the noncompact manifold of Fig. 16.

FIG. 18: The diffusive modes of the geodesic flow on the noncompact manifold of Fig. 16 represented by their cumulative function depicted in the complex plane (Re $F_k$, Im $F_k$) versus the wavenumber $k$. 
Diffusive motion is now possible on this noncompact surface. Figure 17 depicts the position of a typical trajectory measured by the number of boosts \( g_0 \). A central limit theorem rules the number of boosts \( g_0 \) after a long time and this motion has a positive and finite diffusion coefficient.

The hydrodynamic modes of diffusion of this system are constructed by using Eq. (24) with trajectories starting from the center of the Poincaré disk with velocities making an angle \( \theta \) with respect to the horizontal axis. The results are depicted in Fig. 18, which shown the same fractality of the cumulative functions as for the multibaker map and the Lorentz gases.

In conclusion, the theory of the diffusive modes is very general and applies to a broad variety of different dynamical systems.

**IX. ENTROPY PRODUCTION AND IRREVERSIBILITY**

The study of the hydrodynamic modes of diffusion as eigenstates of the Liouvillian operator has shown that these modes are typically given in terms of singular distributions without density function. Since the works by Gelfand, Schwartz, and others in the fifties it is known that such distributions acquire a mathematical meaning if they are evaluated for some test functions belonging to certain classes of functions. The test function used to evaluate a distribution is arbitrary although the distribution is not. Examples of test functions are the indicator functions of the cells of some partition of phase space. In this regard, the singular character of the diffusive modes justifies the introduction of a coarse-graining procedure. This reasoning goes along the need to carry out a coarse graining in order to understand that entropy increases with time, as understood by Gibbs [9] and Poincaré [8] at the beginning of the XXth century.

If a phase-space region \( M_t \) is partitioned into cells \( A \), the probability that the system is found in the cell \( A \) at time \( t \) is given by

\[
\mu_t(A) = \int_A P(x, t) dx
\]

in terms of the probability density \( P(x, t) \) which evolves in time according to Liouville’s equation. If the underlying dynamics has Gibbs’ mixing property, these probabilities converge to their equilibrium value at long times:

\[
\lim_{t \to \pm \infty} \mu_t(A) = \mu_{eq}(A) \quad (47)
\]

The knowledge of the Pollicott-Ruelle resonances \( \pm s_\alpha \) of the forward or backward semigroups allows us to specify the approach to the equilibrium state in terms of the following time asymptotics for \( t \to \pm \infty \):

\[
\mu_t(A) = \mu_{eq}(A) + \sum_\alpha C_\alpha \pm \exp(\pm s_\alpha t) + \cdots \quad (48)
\]

where the coefficients \( C_\alpha \pm \) are calculated using the eigenstates associated with the resonances (see Fig. 5).

The coarse-grained entropy is defined in terms of the probabilities as

\[
S_t(M_t|\{A\}) = -k_B \sum_A \mu_t(A) \ln \mu_t(A) \quad (49)
\]

As a consequence of Gibbs’ mixing property and the decomposition (48), the coarse-grained entropy converges toward its equilibrium value \( S_{eq} \) at long times (see Fig. 19). We notice that the rates of convergence are given by the Pollicott-Ruelle resonances and are thus intrinsic to the system.

For the systems sustaining diffusion, it is possible to prove that the rate of entropy production is the one expected by nonequilibrium thermodynamics. At long times, the phase-space probability density becomes more and more inhomogeneous and acquires the singular character of the diffusive modes which control the long-time evolution. Therefore, the approach of the entropy toward its equilibrium value is determined by the diffusion coefficient. In this way, it has been possible to explicitly calculate the entropy production [2, 3, 6]. The time variation of the coarse-grained entropy over a time interval \( \tau \)

\[
\Delta^\tau S = S_t(M_t|\{A\}) - S_{t-\tau}(M_t|\{A\}) \quad (50)
\]
can be separated into the entropy flow
\[ \Delta \tau S = S_{t\to t+\tau} (\Phi^{-\tau} M(t) \{ A \}) - S_{t\to t+\tau} (M(t) \{ A \}) \] (51)
and the entropy production
\[ \Delta \tau S = \Delta \tau S - \Delta \tau S \] (52)

The entropy production can be calculated using the decomposition of the time evolution in terms of the diffusive modes [2, 3, 6]. The singular character of the diffusive modes implies that the entropy production is not vanishing. Moreover, the entropy production expected from nonequilibrium thermodynamics is obtained:
\[ \Delta \tau S \simeq \tau k_B D \frac{(\text{grad } n)^2}{n} \] (53)
where \( n = \mu_t(M_t) \) is the particle density [2, 3, 6].

According to the forward semigroup, the long-time evolution at positive times is described by the diffusion equation:
\[ \partial_t n \simeq D \partial_x^2 n \quad \text{for} \quad t > 0 \] (54)
In contrast, it is an antidiffusion equation which describes the long-time evolution at negative times according to the backward semigroup:
\[ \partial_t n \simeq -D \partial_x^2 n \quad \text{for} \quad t < 0 \] (55)

Consequently, the entropy increases toward its equilibrium value for both \( t \to \pm \infty \) (see Fig. 19). The fact is that the forward and backward semigroups involve Liouvillian eigenstates which are physically distinct. Indeed, the eigenstates of the forward semigroup are smooth in the unstable phase-space directions but singular in the stable ones and vice versa for those of the backward semigroup. As aforementioned, the splitting of the time evolution into distinct semigroups constitutes a spontaneous breaking of the time-reversal symmetry at the statistical level of description in terms of the Pollicott-Ruelle resonances and antiresonances.

X. CONCLUSIONS

Since Poincaré’s pioneering work, dynamical systems theory has undergone great advances in particular during the last decades with the development of the theory of chaos and fractals. New concepts have been introduced such as the Lyapunov exponents, the Kolmogorov-Sinai entropy per unit time, and the fractal dimensions. Moreover, these concepts have been interconnected by fundamental formulas which concretize the recent progress. Besides, chaotic behavior has been discovered in many systems of statistical mechanics and, in particular, in molecular-dynamics simulations.

These advances have allowed the establishments of new relationships between dynamical systems theory and transport theory in nonequilibrium statistical mechanics. The concept of Pollicott-Ruelle resonance appears to play a fundamental role in order to describe relaxation phenomena and to explain how a mechanism of spontaneous breaking of the time-reversal symmetry can occur in the Liouvillian dynamics.
When the concept of Pollicott-Ruelle resonance is applied to chaotic systems which are spatially periodic and can sustain a transport process of diffusion, it is possible to reconstruct the hydrodynamic modes of diffusion which control the relaxation toward a state of equilibrium. In chaotic systems such as the multibaker map, the Lorentz gases, or the geodesic flow, a state of local microcanonical equilibrium is reached over the collision time scale before the spatial inhomogeneities of the concentration of tracer particles evolves on the long hydrodynamic time scale. This long-time evolution is thus controled by the diffusive modes which can be constructed using the time evolution as a kind of renormalization semigroup. The dispersion relation of diffusion comes out as a Pollicott-Ruelle resonance depending on the wavenumber of the modes. The surprise is that the associated eigenstates are not regular functions but singular distributions with moreover fractal properties. The diffusive modes cannot be represented by their density functions which do not exist. Instead, their cumulative functions exist and depict fractal curves in the complex plane. The Hausdorff dimension of these fractal curves is related to the diffusion coefficient and the Lyapunov exponent at low wavenumbers. This chaos-transport relationship is very similar to the one of the escape-rate formalism [2].

It is remarkable that these new results from dynamical systems theory allows us to establish a connection with nonequilibrium thermodynamics. First of all, the transport coefficient here of diffusion can be directly related to the characteristic quantities of dynamical systems theory. Moreover, the new results lead to an understanding of the way a complex dynamics can induce the increase of the entropy. The singular character of the diffusive modes justifies the use of a coarse-grained entropy and implies that the entropy production is nonvanishing and positive for \( t \rightarrow +\infty \). The thermodynamic considerations are relevant even in systems such as the multibaker map or the Lorentz gases because their mixing dynamics drives the distribution toward a state of local microcanonical equilibrium at intermediate times. Over the long hydrodynamic time, we recover the entropy production expected from nonequilibrium thermodynamics with a direct \textit{ab initio} calculation. The same calculation can be carried out for diffusion in systems with many interacting particles as shown elsewhere [6]. These results can be extended to the other transport processes such as viscosity and heat conductivity.

To summarize, the description of the time evolution in nonequilibrium thermodynamics is based on asymptotic expansions for \( t \rightarrow \pm \infty \) valid for either \( t > 0 \) or \( t < 0 \). The hydrodynamic equations use asymptotic expansions based on Pollicott-Ruelle resonances and other singularities at complex frequencies, which explains the spontaneous breaking of time-reversal symmetry at the level of a description in terms of statistical ensembles. There is here a selection of initial conditions.

The group of time evolution splits into the forward and the backward semigroups which describe the time evolution of statistical ensembles as asymptotic expansions for \( t \rightarrow \pm \infty \). The backward (forward) semigroup may not be prolonged to \( t > 0 \) (\( t < 0 \)) because of the divergence of the asymptotic expansion.

The hydrodynamic modes of diffusion are given by Liouvillian eigenstates which are singular distributions describing the relaxation toward the thermodynamic equilibrium and breaking the time-reversal symmetry. The asymptotic time evolution of the coarse-grained entropy can be calculated by using the hydrodynamic modes and the entropy production of nonequilibrium thermodynamics is recovered. In conclusion, irreversibility is not incompatible with a time-reversal symmetric equation of motion since the solutions of the equation of motion do not need to have the time-reversal symmetry.

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