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Microscopic Chaos and Nonequilibrium Statistical Mechanics: From Quantum to Classical Dynamics

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We review recent results on the relationships between the microscopic chaos in the motion of atoms or molecules in fluids and the transport properties sustained across these macroscopic systems. In the escape-rate formalism, the transport coefficients can be expressed in terms of the positive Lyapunov exponents, the Kolmogorov-Sinai entropy per unit time or the Hausdorff dimension of a fractal repeller generated in the phase space by the nonequilibrium constraints. In the same context, nonequilibrium steady states turn out to be described by singular invariant measures in the large-system limit. This singular character of the nonequilibrium measures explains the entropy production expected from irreversible thermodynamics.

Moreover, we also discuss how the aforementioned classical and kinetic properties can emerge out of the wave dynamics in quantum systems.

I. INTRODUCTION

Today, a dynamical understanding of natural phenomena is under rapid development in nonlinear science and dynamical systems theory. Many recent works have recognized the fact that natural phenomena are essentially temporal and spatio-temporal processes [1]. They are characterized by different time scales given by the rates of instability and of relaxation, which are the Lyapunov exponents and the Pollicott-Ruelle resonances [2, 3]. These new concepts from dynamical systems theory shed a new light on the dynamics of many-body systems which has been studied in nonequilibrium statistical mechanics and thermodynamics since the XIXth century.

During the XXth century, nonequilibrium statistical mechanics has been successfully developed not only on the basis of classical mechanics but also of quantum mechanics. Partly motivated by the study of mesoscopic systems, recent works have focused on dynamical systems with a few to many particles in order to understand how macroscopic behaviours and, in particular, thermodynamical, relaxation and transport processes can be understood as properties emerging out of the mechanics of a particle system [4–6]. In this context, the importance of chaotic behaviour has been emphasized [7, 8] and the hypothesis has been formulated that typical systems of statistical mechanics are chaotic [9, 10]. This chaotic hypothesis replaces the previous stochastic hypothesis – Boltzmann's Stosszahlansatz – and has important consequences which have been recently discovered. In particular, the microscopic chaos implies the formation of fractal structures of trajectories, on which the transport coefficients can be related to the characteristic quantities of chaos such as the Lyapunov exponents, the dynamical entropies and the fractal dimensions [11–15]. Moreover, recent works have shown that irreversible thermodynamics and its entropy production can be understood in terms of the classical chaotic and fractal properties [16]. On the other hand, recent works on complex quantum systems provide us with a scheme to understand how classical chaotic behaviours can in turn emerge out of the fundamental quantum mechanics [17–19].

Our aim is here to give an overview of the theoretical links from quantum to classical and from classical to kinetic properties. We shall also comment on some possible direct connections from quantum to kinetic properties since they are of importance in condensed phases and, specially, in coherent quantum phenomena like superfluidity at low temperatures.

In Section II, we discuss about the emergence of classical and kinetic properties out of quantum mechanics. In Section III, we present recent results on the connection between classical chaos and transport phenomena. In Section IV, we show how irreversible thermodynamics and entropy production have recently been understood in a simple model called the multibaker or bakery map.

II. EMERGENCE OF CLASSICAL AND KINETIC BEHAVIOURS IN QUANTUM SYSTEMS

Quantum dynamics has very peculiar properties due to the linearity of Schrödinger's equation which rules the time evolution of the wavefunction in few-body systems. In thermal many-body systems, the wavefunction description has to be replaced by a description in terms of quantum algebras where the central quantities are the averages and the multiple-time correlation functions of the quantum observables [20]. Indeed, the experimental signal obtained by probing a quantum system is generally given by a time correlation function. For instance, the cross-section of photoabsorption by a molecule or a cluster is given in terms of the auto-correlation function of the electric dipole operator [21]

$$\kappa(\omega) \;=\; \frac{\omega}{6\epsilon_0 c\hbar} \int_{-\infty}^{+\infty} dt\; e^{-i\omega t}\; \langle \hat{\mathbf{D}}(0)\cdot\hat{\mathbf{D}}(t) \;-\; \hat{\mathbf{D}}(t)\cdot\hat{\mathbf{D}}(0)\rangle \;, \tag{1}$$

where $\hbar$ is the Planck constant, c is the light velocity, ϵ_0 is the vacuum permitivity, and $\langle\cdot\rangle$ denotes the average over an initial equilibrium quantum state like the canonical state. On the other hand, the nonlinear optical properties are given in terms of 3-time or 4-time correlation functions [21].
Similarly, the transport coefficients α of diffusion, viscosity, or heat conduction of a thermal many-body system are given in terms of the auto-correlation functions of the associated microscopic current operator $\hat{J}^{(\alpha)}$ as

$$\alpha \;=\; \frac{1}{2}\int_{-\infty}^{+\infty} dt \int_0^{\beta} d\lambda\; \langle \hat{J}^{(\alpha)}(0)\hat{J}^{(\alpha)}(t+i\hbar\lambda)\rangle \;, \qquad (\beta = 1/k_{\rm B}T) \;, \tag{2}$$

as shown by Kubo [22]. If we introduce the corresponding Helfand moment [23]

$$\hat{G}^{(\alpha)}(t) \;=\; \hat{G}^{(\alpha)}(0) \;+\; \int_0^t \hat{J}^{(\alpha)}(\tau)\; d\tau \;, \tag{3}$$

the transport coefficient can be expressed by an Einstein-type formula as

$$\alpha \;=\; \lim_{t\to\infty} \frac{1}{2t} \int_0^{\beta} d\lambda\; \langle|\hat{G}^{(\alpha)}(t+i\hbar\lambda/2) \;-\; \hat{G}^{(\alpha)}(i\hbar\lambda/2)|^2\rangle \;, \tag{4}$$

which proves its non-negativity: $\alpha \geq 0$ (see Appendix).
On the other hand, dynamical randomness is characterized by n-time correlation functions corresponding to the repeated observations of the system at successive times $t_n = n\Delta t$, in order to test the temporal disorder of the dynamics [24–26].
In the previous formulas (2)-(4), the time evolution of the observable is ruled by the system Hamiltonian $\hat{H}$ according to

$$\hat{A}(t) \;=\; \exp\big(+i\hat{H}t/\hbar\big)\; \hat{A}\; \exp\big(-i\hat{H}t/\hbar\big) \;. \tag{5}$$

As a consequence, the dynamical properties of the system are directly determined by the spectrum of its Hamiltonian operator. In this respect, we may distinguish different classes of systems:
1. Few-body systems with a discrete energy spectrum of bound states;
2. Few-body systems with a continuous energy spectrum such as the scattering systems and the spatially periodic systems;
3. Many-body systems with a continuous spectrum and a positive number of particles per unit volume, which are described by quantum algebras [20].

We notice that a photoabsorption cross-section like (1) is composed of Dirac delta peaks centered at the Bohr frequencies $\omega = (E_m - E_n)/\hbar$ for systems with a discrete energy spectrum and is continuous otherwise. In one-particle quantum systems, a transport coefficient like diffusion is typically either infinite ($\alpha = \infty$) in the case of a continuous band-spectrum due to ballistic motion, or zero ($\alpha = 0$) in the case of a discrete spectrum due to localization [27]. Normal transport with $0 < \alpha < \infty$ is therefore expected in many-body quantum systems which are mixing and such that the 2-time correlation functions decay fast enough for their integrals to be finite in eq. (2). This normal transport may be referred to as a kinetic or relaxation property.

Such kinetics properties can be reached in the thermodynamic limit where the number of particles increases with the volume of the system in order to keep constant the particle density. In a N-body system, the average level density typically grows as

$$n_{\rm av}(E) \sim \frac{1}{k_{\rm B}T} \exp\left(\frac{S}{k_{\rm B}}\right) \sim \frac{1}{k_{\rm B}T}\left[\frac{V(mk_{\rm B}T)^{3/2}}{N\ h^3}\right]^N \sim \frac{1}{k_{\rm B}T}\left(\frac{1}{\rho\ \lambda^3_{\rm de\ Broglie}}\right)^N , \tag{6}$$

where $E = 3k_{\rm B}TN/2$, T is the effective temperature, S is the entropy of the system of volume V, m the mass of the particles, $\rho = N/V$ is the particle density, and $h = 2\pi\hbar$ [28]. The wavefunction is characterized by the de Broglie wavelength of the particles

$$\lambda_{\rm de\ Broglie} \equiv \frac{h}{\sqrt{mk_{\rm B}T}} . \tag{7}$$

The physical action of a particle in motion across the volume V is typically

$$\frac{W}{\hbar} \sim \frac{1}{\rho^{1/3}\ \lambda_{\rm de\ Broglie}} , \tag{8}$$

which gives an estimate of the number of nodes of the wavefunction for a particle displaced across a diameter $V^{1/3}$ of the system. As expected, this number is large in a dilute gas at high temperature.

Recent works have shown how the energy levels and the associated eigenfunctions behave in typical discrete spectra as the density of levels increases [18, 19]. The spectrum is known to become irregular due to Wignerian repulsions between the energy levels, while the eigenfunctions display random properties leading to thermalization as shown by Srednicki [29]. Such results justify in particular the use of the canonical ensemble in many-body systems. We may wonder what are the implications on the dynamical properties.

In the case of a discrete energy spectrum, a 2-time correlation function is given by

$$\langle \hat{A}(0)\hat{A}(t)\rangle = \sum_{mn} c_{mn} \exp \frac{i}{\hbar}(E_m - E_n)t , \tag{9}$$

with some coefficients c_{mn}. This function is known to be an almost-periodic function of time due to the discreteness of the spectrum [30]. Its oscillations or quantum beats arise after the Heisenberg time which is proportional to the average level density

$$t_{\rm Heisenberg} \sim \hbar\ n_{\rm av}(E) . \tag{10}$$

The quasiclassical and semiclassical behaviours occur only in the intermediate time scale $0 < t < t_{\rm Heisenberg}$ [31]. Consequently, the kinetic or relaxation phenomena can only exist as emerging properties in systems with an arbitrarily long Heisenberg time. The fact that the Heisenberg time grows exponentially with the entropy and, thus, with the number N of particles shows that the emerging relaxation processes of irreversible thermodynamics could be observed over very long times already in mesoscopic systems. In any cases, we remark that, systems being in contact with the external world at least by the electromagnetic coupling, the assumption of a discrete spectrum is obsolete beyond the time scale of electromagnetic radiative damping.

The Heisenberg time may become arbitrarily large in different limits:

a. In the semiclassical limit $W/\hbar \to \infty$ while N remains finite, we reach Hamilton's equations of classical mechanics and its nonlinear properties of bifurcation and of classical chaos [17–19, 32].

b. In the thermodynamic limit $N, V \to \infty$ and $N/V = \rho$ while keeping constant $W/\hbar$, we reach the thermal states of macroscopic systems. In this limit, the quantum system may have a positive dynamical entropy per unit time characterizing some dynamical randomness [24–26].
c. In other limits, we may reach different mean-field equations like the nonlinear Schrödinger and Hartree-Fock equations of application in condensed phases [33].
We therefore observe that the nonlinear, kinetic and random phenomena appear in some limit $N \to \infty$ or $W/\hbar \to \infty$. In this sense, we may refer to these phenomena as emerging properties of the quantum system. In the following section, we shall assume that the system is in a quasiclassical regime so that it is well described by classical mechanics.

III. CLASSICAL CHAOS

In the quasiclassical limit $W \gg \hbar$, we explained in the previous section that the quantum wavepackets approximately follow the trajectories of classical mechanics. Feynman path integrals show that the dominant contributions to the quantum amplitudes are coming from classical trajectories as given by Hamilton's variational principle based on the action

$$W = \int \mathbf{p} \cdot d\mathbf{q} - H\, dt . \tag{11}$$

The vanishing of its first variation, $\delta W = 0$, leads to Hamilton's equations while its second variation, $\delta^2 W$, provides the properties of the linear stability of the trajectories and, in particular, their Lyapunov exponents [32].
Recent numerical works have shown that classical many-body systems have typically a complete spectrum of positive Lyapunov exponents [6–9, 35, 36]. In a fluid of particles of diameter d, the maximum Lyapunov exponent can be estimated as

$$\lambda_{\rm max} \sim \rho\, d^2 \sqrt{\frac{k_{\rm B}T}{m}}\, \ln\left(\frac{1}{\rho d^3}\right) , \tag{12}$$

which is of the order of the inverse of the time between two successive collisions of a particle. For a gas at room temperature and pressure, we find that $\lambda_{\rm max} \sim 10^{10}\ {\rm sec}^{-1}$. The dynamical randomness of such systems is gigantic because the amount of information required to observe without ambiguity a typical trajectory during a second would be equal to the Kolmogorov-Sinai entropy which is given by

$$h_{\rm KS} = \sum_{\lambda_i > 0} \lambda_i \sim 3\, N\, \lambda_{\rm max} , \tag{13}$$

according to Pesin's formula [2].
Such numerical observations have suggested the hypothesis of microscopic chaos [9, 37, 38] or the chaotic hypothesis [10] that typical systems of statistical mechanics are chaotic. This hypothesis has the advantage of being compatible with deterministic Hamiltonian dynamics in contrast with the previous stochastic hypotheses which assumed an arbitrarily large dynamical randomness incompatible with Newton's equations [38].

IV. RELATIONSHIPS BETWEEN TRANSPORT AND CHAOTIC PROPERTIES

The time scale of microscopic chaos is very short as compared with the hydrodynamic time scales of transport phenomena. For instance, the rate of damping by diffusion of a spatial inhomogeneity of wavelength $L \to \infty$ is arbitrarily small as $\gamma \sim D/L^2$ where D is the diffusion coefficient. Therefore, a relationship between transport and chaos is not a priori evident.
However, dynamical systems theory has shown that nonequilibrium conditions imposed on a chaotic system induce different kinds of fractal objects in phase space [11–15, 39]. These fractal objects like the

fractal repeller of the escape-rate formalism are characterized by fractal dimensions or, equivalently, by a Kolmogorov-Sinai entropy per unit time which differs from the sum of positive Lyapunov exponents under nonequilibrium conditions [12–15]. The difference gives the rate of escape of trajectories out of the fractal repeller as [2]

$$\gamma(\mathcal{R}) \;=\; \sum_{\lambda_i>0} \lambda_i(\mathcal{R}) \;-\; h_{\rm KS}(\mathcal{R}) \; . \tag{14}$$

Recent studies show that the nonequilibrium conditions can be chosen appropriately in order for the escape rate γ to be related to a specific transport coefficient α [14, 15]. This connection is based on the observation that the associated Helfand moment (3) has a diffusive motion, in particular, in the classical limit $\hbar \to 0$. Indeed, eq. (4) shows that the variance of the Helfand moment should grow linearly with time as

$$\langle \big[G^{(\alpha)}(t) - G^{(\alpha)}(0)\big]^2 \rangle \;\simeq\; 2\;\alpha\; k_{\rm B}T\; t \; , \qquad t \to \infty \; . \tag{15}$$

A fractal repeller can be defined by the set of all the trajectories for which the Helfand moment remains forever in the following interval

$$\text{repeller } \mathcal{R} \;: \qquad -\chi/2 \;\leq\; G^{(\alpha)}(t) \;\leq\; +\chi/2 \; . \tag{16}$$

The rate of escape out of this repeller is controlled by the diffusive behaviour and, as a consequence, it is given by

$$\gamma(\mathcal{R}) \;\simeq\; \alpha\; k_{\rm B}T \, \Big(\pi/\chi\Big)^2 \; , \qquad \chi \to \infty \; . \tag{17}$$

Hence, the transport coefficient is given by [12, 14]

$$\alpha \;=\; \lim_{\chi\to\infty} \frac{1}{k_{\rm B}T} \left(\frac{\chi}{\pi}\right)^2 \left[\sum_{\lambda_i>0} \lambda_i(\mathcal{R}) \;-\; h_{\rm KS}(\mathcal{R})\right] . \tag{18}$$

An equivalent formula between the diffusion coefficient and the Hausdorff dimension of the fractal repeller has been derived for systems with two degrees of freedom [13].
Such results show that chaos induces a fractalization in the phase space of nonequilibrium systems. The transport properties turn out to be hidden in the induced fractal objects [13]. Similar fractalizations occur when the nonequilibrium conditions are imposed by quasiperiodic and flux boundary conditions [40, 41].
In particular, the hydrodynamic modes can be constructed with quasiperiodic boundary conditions. In this context, the hydrodynamic modes of diffusion turn out to be the eigenstates of a Frobenius-Perron operator [42–45]. The associated eigenvalues are known as the Pollicott-Ruelle resonances and the dispersion relation of diffusion is given by the dependence of such Pollicott-Ruelle resonances on the wavenumber of the hydrodynamic mode [37, 44]. In this way, the Green-Kubo relation (2) can be derived from first principles in classically chaotic systems of deterministic diffusion [44].

V. NONEQUILIBRIUM STEADY STATES AND ENTROPY PRODUCTION

The previous considerations can be extended to construct the invariant measures describing nonequilibrium steady states as they arise, for instance, in diffusive systems with a gradient g of density [41]. According to recent works [40, 44], such steady states are given in terms of the microscopic current and the associated Helfand moment as

$$\Psi^{(\alpha)}_{\rm st.\;st.}(\mathbf{X}) \;=\; g\left[G^{(\alpha)}(\mathbf{X}) \;+\; \int_0^{-\infty} \; dt\; J^{(\alpha)}(\mathbf{\Phi}^t\mathbf{X})\right] , \tag{19}$$

where $\mathbf{X}$ denotes the positions and momenta of the particles.

In chaotic systems, such a phase-space density defines a singular measure with self-similar properties due to the aforementioned fractalization [41]. This singular character appears when the nonequilibrium conditions are imposed at largely separated boundaries [40]. The chaotic dynamics mixes the trajectories coming from the different boundaries down to very fine scales in phase space. If the nonequilibrium condition consists of injecting particles with green or red colors at the left- or right-hand boundaries respectively, the color can be resolved only below the very fine scales of chaotic mixing. On coarser scales, the particles would be too finely mixed to resolve their color so that the determinism of the process would be lost. The dimension of the very fine scales on which the determinism – i.e., the absolute continuity of the invariant measure – can be resolved decreases exponentially with the size of the system due to the sensitivity to initial conditions of chaos. As a consequence, the nonequilibrium process will behave as predicted by irreversible thermodynamics on any intermediate scale.

In this way, we have been able to derive the positive entropy production [46]

$$\frac{d_i S}{dt} \; = \; D \; \frac{(\text{grad}\ \rho)^2}{\rho} \; + \mathcal{O}\left[\frac{(\text{grad}\ \rho)^4}{\rho^3}\right] , \tag{20}$$

expected from irreversible thermodynamics for a chaotic, volume-preserving model of deterministic diffusion called the multibaker or bakery map [16]. Accordingly, the second law of thermodynamics appears to be a direct consequence of the singular character of the large-system steady state invariant measure (19). The well-known paradox of the constancy of Gibbs' entropy is avoided thanks to this singular character as explained elsewhere [16]. Since chaos is at the origin of the singular character of the steady state we could say that the macroscopic laws of irreversible thermodynamics are the manifestation of the microscopic chaos.

VI. CONCLUSIONS

Modern dynamical systems theory has developed very powerful tools and methods which allow us to understand in detail some of the most fundamental aspects of nonequilibrium statistical mechanics which have remained mysterious and very poorly understood since Boltzmann's pioneering works.

The fact that chaotic behaviour already appears in classical systems with two degrees of freedom provides us with deterministic models which can be analyzed in detail and which show that relaxation phenomena of irreversible thermodynamics are perfectly compatible with a time-reversal symmetric microscopic dynamics. The important condition at the basis of relaxation phenomena is a dynamical instability of the trajectories such as the dynamical instability characterized by positive Lyapunov exponents, as numerically observed in many simulations of molecular dynamics [6–8, 35, 36]. In this perspective, the chaotic hypothesis appears as a fundamental assumption for classical nonequilibrium statistical mechanics. Experimental tests of this hypothesis are possible as explained elsewhere [38].

The extension of the chaotic hypothesis to quantum systems is an important question. Already, in few-body systems, it has recently been suggested that the existence of a Wignerian spectrum of energy levels may be conditioned by some properties of the Pollicott-Ruelle resonances characterizing the relaxation rates of the emerging kinetics [47]. In many-body quantum systems, the characterization of dynamical randomness by dynamical entropies would establish further and stronger justifications for the foundation of quantum statistical mechanics [24–26].

Appendix. According to the definition (3) of a Helfand moment, we find that

$$\begin{aligned} & \langle \big[\hat{G}^{(\alpha)}(t) - \hat{G}^{(\alpha)}(0)\big] \big[\hat{G}^{(\alpha)}(t+i\hbar\lambda) - \hat{G}^{(\alpha)}(i\hbar\lambda)\big] \rangle \\ = & \int_0^t dt' \int_0^t dt'' \; \langle \hat{J}^{(\alpha)}(t')\hat{J}^{(\alpha)}(t''+i\hbar\lambda)\rangle \\ = & \; t \int_{-t}^{+t} d\tau \; (1-|\tau|/t) \; \langle \hat{J}^{(\alpha)}(0)\hat{J}^{(\alpha)}(\tau+i\hbar\lambda)\rangle \; . \end{aligned} \tag{21}$$

For the canonical state, $\langle \cdot \rangle = \text{tr} \exp(-\beta \hat{H})(\cdot)/Z$ with $Z = \text{tr} \exp(-\beta \hat{H})$, and a Hermitian operator $\hat{A} = \hat{A}^\dagger$, we have

$$\langle \hat{A}(0)\hat{A}(i\hbar\lambda)\rangle \; = \; \langle \hat{A}^{\dagger}(i\hbar\lambda/2)\hat{A}(i\hbar\lambda/2)\rangle \; = \; \langle |\hat{A}(i\hbar\lambda/2)|^2\rangle \; , \tag{22}$$

with the definition (5). Performing the integral $\int_0^{\beta} d\lambda$ of (21), dividing both members by $2t$, and taking the limit $t \to \infty$, we obtain (4).

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