Fluctuations in Stationary Nonequilibrium States of Irreversible Processes

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We formulate a dynamical fluctuation theory for stationary nonequilibrium states (SNS) which covers situations in a nonlinear hydrodynamic regime and is verified explicitly in stochastic models of interacting particles. In our theory a crucial role is played by the time reversed dynamics. Our results include the modification of the Onsager-Machlup theory in the SNS, a general Hamilton-Jacobi equation for the macroscopic entropy and a nonequilibrium, nonlinear fluctuation dissipation relation valid for a wide class of systems.

The Boltzmann-Einstein theory of equilibrium thermodynamic fluctuations, as described, for example, in Landau-Lifshitz [1], states that the probability for a fluctuation from equilibrium in a macroscopic region of volume V is proportional to $\exp\{V\Delta S/k\}$, where ΔS is the variation of entropy density calculated along a reversible transformation creating the fluctuation, and k is the Boltzmann constant. This theory is well established and has received a rigorous mathematical formulation in classical equilibrium statistical mechanics via the so-called large deviation theory [2]. The rigorous study of large deviations has been extended to hydrodynamic evolutions of stochastic interacting particle systems [3]. In a dynamical setting, one may ask new questions. For example, what is the most probable trajectory followed by the system in the spontaneous emergence of a fluctuation or in its relaxation to equilibrium? The Onsager-Machlup theory [4] gives the following anwer: In the situation of linear macroscopic equations, that is, close to equilibrium, the most probable emergence and relaxation trajectories are one the time reversal of the other. Developing the methods of [3], this theory has been extended to nonlinear hydrodynamical regimes [5]. Onsager-Machlup assume the reversibility of the microscopic dynamics; however, microscopically nonreversible models were constructed where the above results still hold [6,7].

Emergence of large fluctuations, including Onsager-Machlup symmetry, has been observed in stochastically perturbed gradient-type electronic devices [8]. In their work, these authors study also nongradient (i.e., nonreversible) systems and observe violation of Onsager-Machlup symmetry.

In this Letter, we formulate a general theory of large deviations for irreversible processes, i.e., when a detailed balance condition does not hold. This question was previously addressed in [9]. Natural examples are boundary driven stationary nonequilibrium states (SNS), e.g., a thermodynamic system in contact with two reservoirs, but our theory covers, as a special case, also the model systems considered in [8]. In our approach a crucial role is played by the time reversed dynamics with respect to the stationary nonequilibrium ensemble.

Our results are the following: (i) The Onsager-Machlup relationship has to be modified: The emergence of a fluctuation takes place along a trajectory which is determined by the time reversed process. (ii) We show that the macroscopic entropy solves a Hamilton-Jacobi equation generalizing to a thermodynamic context known results for finite dimensional Langevin equations [10] as those studied in [8]. The Hamilton-Jacobi equation can be solved perturbatively if we consider not too large fluctuations. (iii) We test our theory in a stochastic model of an interacting particles system, the boundary driven zero range process, in which we perform the computations explicitly. In particular, it is possible to construct the microscopic time reversed process and to write the macroscopic entropy in a closed form. (iv) For a large class of systems, we obtain a nonequilibrium nonlinear fluctuation dissipation relationship which links the macroscopic evolution of the system and of its time reversal to the thermodynamic force.

We are interested in a many-body system in the limit of infinitely many degrees of freedom. The basic assumptions of our theory are the following.

(i) The microscopic evolution is given by a Markov process X_t which represents the configuration of the system at time t. This hypothesis probably is not so restrictive because also the Hamiltonian case discussed in [11] in the end is reduced to the analysis of a Markov process. The SNS is described by a stationary, i.e., invariant with respect to time shifts, probability distribution P_{st} over the trajectories of X_t.

(ii) The macroscopic behavior of the system is described by diffusion-type hydrodynamical equations of the form

$$\partial_t \rho = \sum_{i,j}^{d} \partial_{u_i}\{D_{i,j}(\rho)\partial_{u_j}\rho\} = F(\rho), \qquad (1)$$

where $\rho = \rho_t(u)$ represents, in general, a vector of thermodynamic variables, e.g., the densities of different species of particles, and $D_{i,j}$ is a matrix acting on this vector. The interaction with the reservoirs appears as a boundary condition to be imposed on solutions of (1). We assume that there exists a unique stationary solution $\bar{\rho}$ of (1), i.e., a profile $\bar{\rho}(u)$ which satisfies the appropriate boundary conditions such that $F(\bar{\rho}) = 0$.

These equations are derived from the underlying microscopic dynamics through an appropriate scaling limit. The hydrodynamic equations represent laws of large numbers with respect to the probability measure P_{st} conditioned on an initial state X_0. The initial conditions for (1) are determined by X_0. Of course, many microscopic configurations give rise to the same value of $\rho_0(u)$. In general, $\rho_t(u)$ is an appropriate limit of a $\rho_N(X_t)$ as the number N of degrees of freedom diverges.

(iii) The stationary measure P_{st} admits a principle of large deviations describing the fluctuations of the thermodynamic variables appearing in the hydrodynamic equations. This means the following. The probability that in a macroscopic volume V containing N particles the evolution of the variable ρ_N deviates from the solutions of the hydrodynamic equations, and is close to some trajectory $\hat{\rho}_t$, is exponentially small and of the form

$$P_{\mathrm{st}}\{\rho_N(X_t) \approx \hat{\rho}_t(u), t \in [t_1, t_2]\} \approx e^{-N[S(\hat{\rho}_{t_1}) + J(\hat{\rho})]}$$
$$= e^{-NI(\hat{\rho})}, \qquad (2)$$

where $J(\hat{\rho})$ is a functional, called the action functional, which vanishes if $\hat{\rho}_t$ is a solution of (1), and $S(\hat{\rho}_{t_1})$ is the entropy cost to produce the initial value $\hat{\rho}_{t_1}$. We adopt the convention for the entropy sign opposite to the usual one, so that it takes the minimum value in the equilibrium state. We also normalize it so that $S(\bar{\rho}) = 0$. Therefore $J(\hat{\rho})$ represents the extra cost necessary in order that the system follow the trajectory $\hat{\rho}_t$. Finally, $\rho_N(X_t) \approx \hat{\rho}_t(u)$ means closeness in some metric. This formula is a generalization of the Boltzmann-Einstein formula in which we set the Boltzmann constant $k = 1$.

(iv) Let us denote by θ the time inversion operator defined by $\theta X_t = X_{-t} = X_t^*$. The probability measure P_{st}^* describing the evolution of the time reversed process X_t^* is given by the composition of P_{st} and θ^{-1}; that is,

$$P_{\mathrm{st}}^*(X_t^* = \phi_t, t \in [t_1, t_2]) = P_{\mathrm{st}}(X_t = \phi_{-t},$$
$$t \in [-t_2, -t_1]). \qquad (3)$$

Let L be the generator of the microscopic dynamics. We remind that L induces the evolution of functions of the process according to the equation $\partial_t E[f(X_t)] = E[(Lf)(X_t)]$, where E stands for the expectation with respect to P_{st} conditioned on the initial state X_0 [12]. The time reversed dynamics is generated by the adjoint L^* of L with respect to the invariant measure

P_{in}; that is, $E^{P_{\mathrm{in}}}[fLg] = E^{P_{\mathrm{in}}}[(L^*f)g]$. The measure P_{in}, which is the same for both processes, is a distribution over the configurations of the system and formally satisfies $P_{\mathrm{in}}L = 0$. $E^{P_{\mathrm{in}}}$ is the expectation with respect to P_{in} and f, g are functions of the configuration. We require that also the evolution generated by L^* admits a hydrodynamic description, which we call the adjoint hydrodynamics, which, however, is not necessarily of the same form as (1). In fact, the adjoint hydrodynamics can be nonlocal in space.

In order to avoid confusion we emphasize that what is usually called an equilibrium state, as distinguished from a SNS, corresponds to the special case $L^* = L$, i.e., the detailed balance principle holds. In such a case, P_{st} is invariant under time reversal and the two hydrodynamics coincide.

We now derive a fundamental consequence of our assumptions, that is, the relationship between the functionals I and I^* associated to the dynamics L and L^*. From Eq. (3) and our assumptions, it follows that also P_{st}^* admits a large deviation principle with functional I^* given by

$$I_{[t_1,t_2]}^*(\hat{\rho}) = I_{[-t_2,-t_1]}(\theta\hat{\rho}), \qquad (4)$$

with obvious notations. More explicitly this equation reads

$$S(\hat{\rho}_{t_1}) + J_{[t_1,t_2]}^*(\hat{\rho}) = S(\hat{\rho}_{t_2}) + J_{[-t_2,-t_1]}(\theta\hat{\rho}), \qquad (5)$$

where $\hat{\rho}_{t_1}, \hat{\rho}_{t_2}$ are the initial and final points of the trajectory and $S(\hat{\rho}_{t_i})$ the entropies associated with the creation of the fluctuations $\hat{\rho}_{t_i}$ starting from the SNS. The functional J^* vanishes on the solutions of the adjoint hydrodynamics. To compute J^* it is necessary to know the macroscopic entropy S, which is determined by the microscopic invariant measure P_{in}. We shall see that S can be also obtained as the solution of a Hamilton-Jacobi equation involving only macroscopic quantities. From (5) we can already obtain the generalization of the Onsager-Machlup relationship for SNS.

The physical situation we are considering is the following. The system is macroscopically in the stationary state $\bar{\rho}$ at $t = -\infty$, but at $t = 0$ we find it in the state $\hat{\rho}_0$. We want to determine the most probable trajectory followed in the spontaneous creation of this fluctuation. According to (2) this trajectory is the one that minimizes J among all trajectories connecting $\bar{\rho}$ to $\hat{\rho}_0$ in the time interval $[-\infty, 0]$. From (5) we have

$$J_{[-\infty,0]}(\hat{\rho}) = J_{[0,\infty]}^*(\theta\hat{\rho}) + S(\hat{\rho}_0). \qquad (6)$$

The right-hand side is minimal if $J_{[0,\infty]}^*(\theta\hat{\rho}) = 0$, that is, if $\theta\hat{\rho}$ is a solution of the adjoint hydrodynamics. The existence of such a relaxational solution is due to the fact that the stationary solution $\bar{\rho}$ is attractive also for the adjoint hydrodynamics. We have therefore the following generalization of Onsager-Machlup to SNS.

In a SNS, the spontaneous emergence of a macroscopic fluctuation takes place most likely following a trajectory which is the time reversal of the relaxation path according to the adjoint hydrodynamics.

The form of the action functional J is given by the large fluctuation theory (see, e.g., [13]) and depends on the model considered; we assume that J is a quadratic form,

$$J_{[t_1,t_2]}(\hat{\rho}) = \frac{1}{2} \int_{t_1}^{t_2} dt \langle W, K(\hat{\rho})W \rangle, \quad (7)$$

where $W = \partial_t \hat{\rho} - F(\hat{\rho})$, $F(\hat{\rho})$ has been defined in (1), $\langle \cdot, \cdot \rangle$ denotes integration in the space variable, and $K(\hat{\rho})$ is a positive kernel. This is the form of J we expect for lattice gases. It is also typical for finite dimensional diffusion processes [10] where K^{-1} is the diffusion matrix in the Fokker-Planck equation. In general, K reflects, at the macroscopic level, the stochasticity of the system.

From (6) we have that the entropy is related to J by

$$S(\rho) = \inf_{\hat{\rho}} J_{[-\infty,0]}(\hat{\rho}), \quad (8)$$

where the minimum is taken over all trajectories connecting $\bar{\rho}$ to ρ. Therefore S must satisfy the Hamilton-Jacobi equation associated to the action functional J. A simple calculation gives

$$\frac{1}{2} \left\langle \frac{\delta S}{\delta \rho}, K^{-1}(\rho) \frac{\delta S}{\delta \rho} \right\rangle + \left\langle \frac{\delta S}{\delta \rho}, F(\rho) \right\rangle = 0. \quad (9)$$

One can try to solve this functional derivative equation by successive approximations. Let $\bar{\rho}(u)$ be the stationary profile in the SNS. If we expand S as a Volterra functional series in the argument $\rho - \bar{\rho}$, Eq. (9) reduces to a system of equations for the kernels of the expansion which can be solved by iteration. Applications of (9) to various models will be discussed in [14].

We consider now the so-called zero-range process which models a nonlinear diffusion of a lattice gas (see, e.g., [13]). The model is described by a positive integer variable $\eta_\tau(x)$ representing the number of particles at site x and time τ of a finite lattice which for simplicity we assume is one dimensional. The particles jump with rates $g(\eta(x))$ to one of the nearest-neighbor sites $x + 1, x - 1$ with probability $1/2$. The function $g(k)$ is nondecreasing and $g(0) = 0$. We assume that our system interacts with two reservoirs of particles in positions N and $-N$ with rates p_+ and p_-, respectively. The microscopic dynamics is then defined by the generator [15],

$$(L_N f)(\eta) = \frac{1}{2} \sum_{x=-N}^{N} g(\eta(x))[\nabla^{x,x+1}f + \nabla^{x,x-1}f]$$

$$+ \frac{1}{2} p_+[f(\eta^N) - f(\eta)]$$

$$+ \frac{1}{2} p_-[f(\eta^{-N}) - f(\eta)], \quad (10)$$

where $\nabla^{x,x\pm1}f = f(\eta^{x,x\pm1}) - f(\eta)$ and

$$\eta^{x,y}(z) = \begin{cases} \eta(z) & \text{if } z \neq x, y \\ \eta(z) - 1 & \text{if } z = x \\ \eta(z) + 1 & \text{if } z = y, \end{cases}$$

that is, $\eta^{x,y}$ is the configuration obtained from η when a particle jumps from x to y. Similarly $\eta^{N,N+1}(N) = \eta(N) - 1$, $\eta^{-N,-N-1}(-N) = \eta(-N) - 1$, and $\eta^N(\eta^{-N})$ is the configuration η after addition of a particle at the point $N(-N)$.

It is remarkable that the invariant measure for this process can be constructed explicitly [15]. It is the grand-canonical measure $P_N(\eta)$ obtained by taking the product of the marginal distributions,

$$P_N[\eta(x) = k] = \frac{\lambda_N^k(x)}{g(1) \cdots g(k)} Z_N^{-1}(x), \quad (11)$$

where

$$\lambda_N(x) = \frac{p_+ - p_-}{2(N + 1)} x + \frac{p_+ + p_-}{2}, \quad (12)$$

and

$$Z_N(x) = 1 + \sum_{k=1}^{\infty} \frac{\lambda_N^k(x)}{g(1) \cdots g(k)}. \quad (13)$$

We emphasize that, if $p_- \neq p_+$, the generator L_N is not self-adjoint with respect to the invariant measure so that the process is different from its time reversal and detailed balance does not hold.

Let us introduce now the macroscopic time $t = \tau/N^2$ and space $u = x/N$ and the empirical density,

$$\rho_N(t, u) = \frac{1}{N} \sum_{x=-N}^{N} \eta_{N^2 t}(x) \delta(u - x/N), \quad (14)$$

where $\delta(u - x/N)$ is the Dirac δ.

We prove in [14], adapting the methods of [13], that in the limit $N \to \infty$ the empirical density (14) tends in probability to a continuous function $\rho_t(u)$ which satisfies the following hydrodynamic equation

$$\partial_t \rho = \partial_u \{D(\rho) \partial_u \rho\} = \frac{1}{2} \partial_u^2 \phi(\rho) = F(\rho), \quad (15)$$

where $\phi(\rho)$ is the inverse function of

$$\rho(\phi) = \frac{1}{Z(\phi)} \sum_{k=0}^{\infty} \frac{\phi^k}{g(1) \cdots g(k)} k, \quad (16)$$

where $Z(\phi)$ is the normalization constant as in (13). The function ϕ is well defined because the right-hand side of (16) is strictly increasing in $\phi \geq 0$. Then $\rho(\phi)$ is the equilibrium density corresponding to the activity ϕ. The boundary conditions for (15) are $\phi[\rho_t(\pm1)] = p_\pm$.

From the knowledge of the invariant measure, one can calculate the adjoint generator L_N^* given by

$$(L_N^* f)(\eta) = \frac{1}{2} \sum_{x=-N}^{N} g(\eta(x)) \frac{\lambda_N(x + 1)}{\lambda_N(x)} \nabla^{x,x+1}f(\eta) + \frac{1}{2} \sum_{x=-N}^{N} g(\eta(x)) \frac{\lambda_N(x - 1)}{\lambda_N(x)} \nabla^{x,x-1}f(\eta)$$

$$+ \frac{1}{2} \lambda_N(N)[f(\eta^N) - f(\eta)] + \frac{1}{2} \lambda_N(-N)[f(\eta^{-N}) - f(\eta)]. \quad (17)$$

Notice that the form of (17) is the same as (10) with the rates modified in such a way to invert the particle flux. From (17), one can derive [14] the adjoint hydrodynamics

$$\partial_t \rho = \frac{1}{2} \partial_u^2 \phi(\rho) - \alpha \partial_u \left\{ \frac{\phi(\rho)}{\lambda(u)} \right\} = F^*(\rho), \quad (18)$$

with $\lambda(u) = \frac{p_+ - p_-}{2} u + \frac{p_+ + p_-}{2}$ and $\alpha = \frac{p_+ - p_-}{2}$. The boundary conditions for (18) are the same as for (15).

The second term on the right-hand side of (18) is produced by the new rates in (17). As expected, it is proportional to the difference of the chemical potentials.

The action functionals $J(\hat{\rho})$ and $J^*(\hat{\rho})$ for this model have been computed with the methods of [13]. The basic idea is to add an external space time dependent field such that $\hat{\rho}$ becomes a hydrodynamic trajectory; the details will be given in [14]. The results are

$$J_{[t_1,t_2]}(\hat{\rho}) = \frac{1}{2} \int_{t_1}^{t_2} dt \left\langle \nabla_u^{-1} W, \frac{1}{\phi(\hat{\rho})} \nabla_u^{-1} W \right\rangle, \quad (19)$$

$$J_{[t_1,t_2]}^*(\hat{\rho}) = \frac{1}{2} \int_{t_1}^{t_2} dt \left\langle \nabla_u^{-1} W^*, \frac{1}{\phi(\hat{\rho})} \nabla_u^{-1} W^* \right\rangle, \quad (20)$$

with W as in (7) and $W^* = \partial_t \hat{\rho} - F^*(\hat{\rho})$. Moreover, J and J^* are defined to be infinite if $\hat{\rho}_t$ does not satisfy the boundary conditions stated above.

The entropy $S(\rho)$ can be easily computed from the expression (11) for the invariant measure,

$$S(\rho) = \int_{-1}^{1} du \left[\rho(u) \log \frac{\phi[\rho(u)]}{\lambda(u)} - \log \frac{Z\{\phi[\rho(u)]\}}{Z[\lambda(u)]} \right]. \quad (21)$$

Indeed $S(\rho)$ is obtained as the Legendre transform of the pressure given by

$$G(\mu) = \lim_{N \to \infty} \frac{1}{N} \log \sum_{\eta} P_N(\eta) e^{\sum_x \mu(x/N)\eta(x)}.$$

From Eq. (19), (20), and (21), one can verify explicitly Eq. (5) and the generalized Onsager-Machlup relationship.

We deduce now a twofold generalization of the celebrated fluctuation-dissipation relationship: It is valid in nonequilibrium states and in nonlinear regimes. Such a relationship will hold, provided the rate function $J_{[t_1,t_2]}^*$ of the time reversed process is of the form (7) with the same metric $K(\rho)$ but a different vector field F^* describing the hydrodynamic of the adjoint process, namely,

$$J_{[t_1,t_2]}^*(\hat{\rho}) = \frac{1}{2} \int_{t_1}^{t_2} dt \langle W^*, K(\hat{\rho}) W^* \rangle. \quad (22)$$

By taking the variation of Eq. (5), a simple computation gives

$$F(\rho) + F^*(\rho) = -K(\rho)^{-1} \frac{\delta S}{\delta \rho}. \quad (23)$$

This relation holds for the nonequilibrium zero-range model discussed previously. We also note that it holds for the equilibrium reversible models for which the large deviation principle has been rigorously proven such as the simple exclusion process [3] and the Landau-Ginzburg model [16] and its nongradient version [17]. It is also easy to check that the linearization of (23) around the stationary profile $\bar{\rho}$ yields a fluctuation-dissipation relationship which in equilibrium reduces to the usual one.

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