Le Chatelier Principle for Out-of-Equilibrium and Boundary-Driven Systems: Application to Dynamical Phase Transitions

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A stability analysis is presented for boundary-driven and out-of-equilibrium systems in the framework of the hydrodynamic macroscopic fluctuation theory. A Hamiltonian description is proposed which allows us to thermodynamically interpret the additivity principle. A necessary and sufficient condition for the validity of the additivity principle is obtained as an extension of the Le Chatelier principle. These stability conditions result from a diagonal quadratic form obtained using the cumulant generating function. This approach allows us to provide a proof for the stability of the weakly asymmetric exclusion process and to reduce the search for stability to the solution of two coupled linear ordinary differential equations instead of nonlinear partial differential equations. Additional potential applications of these results are discussed in the realm of classical and quantum systems.

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Understanding out-of-equilibrium systems is an essential problem in physics [1] but surprisingly enough, it still lacks both a macroscopic approach comparable to thermodynamics and a microscopic theory. However, a fruitful hydrodynamic description of driven diffusive systems far from equilibrium, the macroscopic fluctuation theory (MFT), has been proposed [2]. It is based on a variational principle that provides equations for the time evolution of the most probable density profile corresponding to a given fluctuation. The MFT was used to explore aspects of out-of-equilibrium systems [3–8]. The case of current fluctuations has been singled out due to its relevance to problems generically known as full counting statistics, which play an important role both in classical and quantum systems [9–13]. Quite often, a classical description is convenient enough to account for the behavior of quantum systems driven out of equilibrium [14–18]. A great amount of effort has been devoted to the investigation of large current fluctuations since they provide a measure of the likeliness of the system to return to equilibrium. Current fluctuations close to the steady state are expected to be time independent, but far away, the system may choose a time-dependent fluctuation, very much like a phase transition.

To make these considerations more precise, we consider a large system of size L connected to reservoirs of particles at different densities. The system reaches a nonequilibrium steady state with a fluctuating particle current. These fluctuations are characterized by the probability $P_t(Q)$ for having Q particles flowing through the system during a time t. In the long time limit, this probability follows a large deviation principle [19,20],

$$\frac{1}{t}\log P_t(Q) \equiv -\frac{1}{L}\Phi(Q/t). \tag{1}$$

Finding an explicit expression for the large deviation function Φ is a difficult optimization problem. However, a useful and elegant additivity principle (AP) has been formulated [21], which, by assuming that the optimal current trajectory is time independent, reduces the calculation of Φ to solving an Euler-Lagrange equation. A breakdown of the AP signals the onset of a dynamical phase transition. One purpose of this Letter is to formulate a necessary and sufficient condition for the validity of the AP for boundary-driven systems with and without a uniform external field E. This will extend previous results [22–24] and allow us to discuss the existence and nature of such transitions.

Although out-of-equilibrium physics requires new approaches, different from the familiar thermodynamic concepts, it is intuitively helpful to relate these two situations. A powerful idea to study systems at thermodynamic equilibrium is provided by the Le Chatelier principle, which states that the net outcome of a fluctuation is to bring the system back to equilibrium, or, stated otherwise, thermodynamic potentials are concave (convex) functions. It is possible, using the Onsager relations, to extend the Le Chatelier principle to systems out of equilibrium. To that purpose, we recall that a system brought slightly out of equilibrium by the application of forces X_i such as temperature or density gradients behaves diffusively and creates fluxes $\mathcal{J}_i$ linearly related to the forces, $\mathcal{J}_i = \sum_j L_{ij} X_j$. Forces and their related fluxes are such that the products $\mathcal{J}_i X_i$ are additive terms in the corresponding entropy creation. A generalization of the Le Chatelier principle is obtained from the expression $\mathfrak{s} = \sum_i \mathcal{J}_i X_i$ of the entropy creation per unit time. Thus, using the definition of the $\mathcal{J}_i$'s and the symmetry of the L_{ij}'s leads to the positive quadratic form

$$\mathfrak{s} = \sum_{ij} L_{ij} X_i X_j, \tag{2}$$

which implies that $L_{mm} \geq 0$. Then, varying the force X_m by δX_m, we obtain from Eq. (2) that $\mathcal{J}_m \delta X_m \geq 0$; namely, the

flux and the fluctuation generating it are always of the same sign, so that the response of the system tends to act against the perturbation. This is the content of the Le Chatelier principle for nonequilibrium and its breakdown signals the possible onset of a phase transition.

We wish now to implement these ideas using the MFT. To that purpose and for the sake of simplicity, we restrict our study to one-dimensional systems although generalizations to higher dimensions have been proposed [25]. We consider a lattice gas such that $n_i(t)$, $i \in 1, \ldots, L$ denote the time-dependent occupancies of the $L \gg 1$ sites of the system coupled to two reservoirs at its endpoints. The MFT relies on the replacement of the dynamics of the system (either deterministic or not) by a stochastic hydrodynamic equation that describes correctly the fluctuations of the driven system in the long time and large size limits. The relevant physical quantities are the density $\rho(x,\tau)$ and the current density $j(x,\tau)$ of a fluctuating diffusive system, with the scaling $x = i/L$ and $\tau = t/L^2$. The boundary conditions for the density are fixed by $\rho_{l,r}$ at the left and right boundaries $x = 0$, 1. The evolution of the system in the presence of an external field E is described by a Langevin equation

$$j(x,\tau) = -D(\rho)\partial_x\rho + E\sigma(\rho) + \sqrt{\sigma(\rho)}\eta(x,\tau), \quad (3)$$

together with the continuity equation $\partial_\tau\rho = -\partial_x j$. The term $\eta(x,\tau)$ is a multiplicative white noise with zero mean and variance $(1/L)\delta(x-x')\delta(\tau-\tau')$. The phenomenological diffusion $D(\rho)$ and conductivity (transport) $\sigma(\rho)$ coefficients may be obtained from the details of the microscopic process. On average, the current is determined by Fick's law and by a term proportional to the applied field E (linear response to a weak field). The strength $\sigma(\rho)$ of the noise term (dissipative conductivity) is related to the Fick's term by means of an Einstein relation just as in the equilibrium fluctuation-dissipation relation [26]. This generalizes the usual Langevin equation where the strength of the stochastic noise is driven by temperature only.

The number of particles Q in Eq. (1) is the integral of the current density

$$Q = L^2 \int_0^1 dx \int_0^{t/L^2} d\tau j(x,\tau). \quad (4)$$

The two coefficients $D(\rho)$ and $\sigma(\rho)$ can be expressed using the first two cumulants of the probability $P_t(Q)$. To further establish these expressions, we consider now the case $E = 0$ in Eq. (3). In the limit $\rho_R - \rho_L = \Delta\rho \ll 1$ of a slightly out-of-equilibrium system, the steady state average current $\langle Q\rangle/t$ is obtained from Eq. (4) and given by $\langle Q\rangle/t = -(1/L)D(\rho)\Delta\rho$. For $\Delta\rho \to 0$, the variance of the integrated current is $\langle Q^2\rangle_C/t = (1/L)\sigma(\rho)$.

The probability $P_t(Q,\rho_L,\rho_R)$ is obtained in this framework using a stochastic path integral representation [9,27,28]

$$P_t(\{j,\rho\}) \sim \exp\left(-L\int_0^{t/L^2} d\tau \int_0^1 dx\mathcal{L}\right), \quad (5)$$

corresponding to a set $\{j(x,\tau),\rho(x,\tau)\}$ of current and density trajectories. The Lagrangian density $\mathcal{L}(\rho,\partial_x\rho)$ is

$$\mathcal{L} = \frac{[j + D(\rho)\partial_x\rho]^2}{2\sigma(\rho)}, \quad (6)$$

and a saddle point approximation for large L allows us to rewrite the large deviation function in Eq. (1) as [22]

$$\Phi\left(\frac{Q}{t}\right) = \frac{L^2}{t}\inf_{j,\rho}\int_0^{t/L^2} d\tau \int_0^1 dx\mathcal{L}, \quad (7)$$

where the minimum is over all $\rho(x,\tau)$ and $j(x,\tau)$ profiles defined in the time interval $0 < \tau < t/L^2$ and which satisfy the continuity equation and relation (4). The hard minimization problem of finding the optimal current trajectory $j(x,\tau)$ greatly simplifies by assuming the optimal current to be constant, $j(x,\tau) = J$ (up to a macroscopically negligible transient), so that Eq. (4) rewrites $Q/t = J$. This assumption introduced in Ref. [21] is known as the AP. A spatially constant current implies, through the continuity equation, a stationary density $\rho(x)$, so that the corresponding Lagrangian density obtained from Eq. (6) and now denoted $\mathcal{L}_J$ becomes time independent. Therefore, the AP amounts to replacing $\Phi(J)$ in Eq. (7) by

$$U(J) = \inf_{\rho(x)}\int_0^1 dx\mathcal{L}_J(\rho(x),\partial_x\rho(x)). \quad (8)$$

Note that the time variable in the usual Lagrangian description is replaced here by the spatial coordinate. The approximate large deviation function $U(J)$ and the "trajectory" of $\rho(x)$ for the stationary density profile under the AP assumption are then obtained from the associated Euler-Lagrange equation $(d/dx)(\delta\mathcal{L}_J/\delta\partial_x\rho) = (\delta\mathcal{L}_J/\delta\rho)$. It is useful to look at the equivalent Hamiltonian formalism, where the corresponding Hamiltonian is [29]

$$H(\mathcal{P},q) = \frac{1}{2m(q)}[\mathcal{P} - eA(q)]^2 + V(q) \quad (9)$$

with the definitions $q = \rho$ and $\mathcal{P} = \frac{\delta\mathcal{L}}{\delta\partial_x\rho}$ for the conjugate momentum. The Hamiltonian (9) describes a single particle of q-dependent mass $m(q) = D^2(q)/\sigma(q)$ and of charge $e = J$ placed in scalar $V(q) = -e^2/2\sigma(q)$ and "vector" $A(q) = D(q)/\sigma(q)$ potentials. As just stressed, space replaces time; namely, time conservation in Hamiltonian systems translates here into a conservation in space of the associated energy $H(\mathcal{P},q)$, so that the energy is spatially uniform. This provides a useful analogy with thermodynamics, where, at equilibrium, the total energy is uniformly distributed in

space. Therefore, the AP provides, for out-of-equilibrium systems, the analog of a thermodynamic description.

A careful study of the conditions under which the AP is valid thus appears to be essential, since a breakdown of the AP may signal the onset of a (dynamical) phase transition. This question has been investigated in Ref. [22] for closed systems with periodic boundary conditions $\rho(0,\tau) = \rho(1,\tau)$, and a sufficient and necessary condition for the validity of the AP has been given. However, in that case, periodic boundary conditions and particle conservation greatly simplify the problem. Here, we wish to provide a necessary and sufficient condition for the validity of the AP in boundary-driven systems. This question has been addressed using a direct stability analysis of the large deviation function against time dependent perturbations [30], but without a conclusive formulation of a validity criterion for the AP. Although the large deviation function is usually considered to study the stability of the AP solution, we find it far more convenient to work with its Legendre transform

$$\mu(\lambda) = -\frac{1}{L}\inf_J\{\Phi(J) - \lambda J\} = \frac{1}{t}\ln\langle e^{\lambda Q/L}\rangle, \quad (10)$$

since this choice allows to relax the continuity equation constraint and to reformulate Eq. (4) as a boundary condition [31,32]. The notation $\langle\cdot\rangle$ stands for averaging with respect to $P_t(Q)$ given in Eq. (1). Being cautious about the corresponding change of boundary conditions, it is possible to relate $\mu(\lambda)$ to the MFT description by

$$\langle e^{\lambda Q/L}\rangle = \int \mathcal{D}q\mathcal{D}p \exp\left[-L\int dx d\tau S(x,\tau)\right], \quad (11)$$

where q stands for the density, p is a Lagrange multiplier associated with the continuity equation [32], and the action $S(x,\tau)$ is given by

$$S(x,\tau) = D\partial_x q\partial_x p - \frac{\sigma}{2}(\partial_x p)^2 + (p-\lambda x)\partial_\tau q. \quad (12)$$

The corresponding equations of motion can be readily obtained from $\delta S/\delta q = \delta S/\delta p = 0$ [33]

$$\begin{aligned}\partial_\tau q &= \partial_x(D\partial_x q) - \partial_x(\sigma\partial_x p),\\ \partial_\tau p &= -D\partial_{xx} p - \frac{\sigma'}{2}(\partial_x p)^2,\end{aligned} \quad (13)$$

where the notations (D',σ') stand for derivatives with respect to the density q. Now, we consider the AP, which assumes a time-independent density and momentum, so that taking $\partial_\tau q = \partial_\tau p = 0$ the AP equations of motion become two ordinary differential equations for the corresponding (q_0, p_0) with the time-independent boundary conditions

$$\begin{aligned}q(0,\tau) &= \rho_l,\\ q(1,\tau) &= \rho_r,\\ p(0,\tau) &= 0,\\ p(1,\tau) &= -\lambda.\end{aligned} \quad (14)$$

The most probable density profile under the AP is obtained by solving these Hamilton-Jacobi equations with boundary conditions (14).

To discuss the stability of the AP solution, we consider the effect of time-dependent fluctuations $\delta q(x,\tau)$ and $\delta p(x,\tau)$ on the extremum solution (q_0, p_0) and we calculate the variation δS^2_{AP} of the action (12) up to second order in $(\delta q, \delta p)$ [34].

Using a general result for the stability of Hamiltonian systems, the variation δS^2_{AP}, can be written as the diagonal quadratic form [32,35]

$$\delta S^2_{\mathrm{AP}}(x,\tau) = -\frac{D'\sigma' - \sigma'' D}{4D}(\partial_x p_0)^2\delta q^2 - \frac{\sigma}{2}(\partial_x \delta p)^2. \quad (15)$$

This expression constitutes one of the main results of this Letter. It could not be obtained or even anticipated [32] using the large deviation function formalism. The diagonal quadratic form makes it easy to discuss the validity of the AP approximation, which requires $\int dx d\tau \delta S^2_{\mathrm{AP}} < 0$, a condition equivalent to the Le Chatelier condition (2). Noting that σ and D are non-negative (for any q) then, having

$$D'\sigma' \geq \sigma'' D \quad (16)$$

in Eq. (15) implies $\int dx d\tau \delta S^2_{\mathrm{AP}} \leq 0$ for any fluctuation $(\delta q, \delta p)$. Therefore, Eq. (16) is a sufficient condition for the validity of the AP solution. An apparently related but much more stringent condition has been obtained by Bertini *et al.* [22]. Indeed, whereas Eq. (16) needs to be only valid for the density profile $q_0 = \rho_{\mathrm{AP}}$, the related condition in Ref. [22] must be verified for all values of q [36]. However, since the variations δq and δp are not independent but related by (the conveniently linearized) Eqs. (13), it is clear that Eq. (16) is not a necessary condition for stability. Stated otherwise, the AP solution becomes unstable if and only if there exists a fluctuation $(\delta q, \delta p)$ such that $\int dx d\tau \delta S^2_{\mathrm{AP}} > 0$.

To find a necessary and sufficient condition for the stability of the AP solution, we now consider the Fourier spectrum of the time-dependent fluctuations $\delta q(x,\tau)$ and $\delta p(x,\tau)$. Since time is defined on $[0,T]$ where $T = t/L^2$, these fluctuations admit the Fourier series expansion $\delta q = \sum_\omega e^{i\omega\tau} f_\omega(x)$ and $\delta p = \sum_\omega e^{i\omega\tau} g_\omega(x)$ with discrete frequencies $\omega_m = (2\pi/T)m$, $(m \in \mathbb{Z})$. The linearization of Eqs. (13) used to obtain the quadratic form (15) together with having real valued fluctuations lead for the Fourier amplitudes to the set of coupled differential linear equations [35]

$$i\omega f_\omega = \partial_x(D'\partial_x q_0 f_\omega + D\partial_x f_\omega - \sigma'(\partial_x p_0) f_\omega - \sigma\partial_x g_\omega),$$
$$i\omega g_\omega = \left[-D'\partial_{xx}p_0 - \frac{\sigma''}{2}(\partial_x p_0)^2\right] f_\omega - D\partial_{xx} g_\omega - \sigma'\partial_x p_0 \partial_x g_\omega, \quad (17)$$

which, using the equalities $\int d\tau \delta q^2 = \sum_{\omega>0}|f_\omega|^2$ and $\int d\tau(\partial_x \delta p)^2 = \sum_{\omega>0}|\partial_x g_\omega|^2$, allows us to rewrite the fluctuation (15) of the action as $\int dx d\tau \delta S^2_{\mathrm{AP}}(x,\tau) = -\sum_{\omega>0}\delta s^2_\omega$, where [35]

$$\delta s^2_\omega \equiv \int dx \frac{D'\sigma' - D\sigma''}{4D}(\partial_x p_0)^2 |f_\omega(x)|^2 + \frac{\sigma}{2}|\partial_x g_\omega(x)|^2. \quad (18)$$

The AP is stable if and only if $\delta s^2_\omega \geq 0$ for any solution (f_ω, g_ω) of Eq. (17) and $\forall \omega > 0$. To prove this statement, we first assume that $\delta s^2_\omega \geq 0$ for any solution (f_ω, g_ω) and $\forall \omega > 0$. Then, necessarily $\sum_{\omega>0}\delta s^2_\omega \geq 0$ and $\int dx d\tau \delta S^2_{\mathrm{AP}} \leq 0$ for any solution $(\delta q, \delta p)$ so that the AP is stable. Conversely, if there exists a mode ω_0 such that for the solution $(f_{\omega_0}, g_{\omega_0})$ of Eq. (17) $\delta s^2_{\omega_0} < 0$, then one can choose $\delta q = e^{i\omega_0\tau} f_{\omega_0} + e^{-i\omega_0\tau} f^\star_{\omega_0}$ and $\delta p = e^{i\omega_0\tau} g_{\omega_0} + e^{-i\omega_0\tau} g^\star_{\omega_0}$ so that $\delta s^2_\omega = 0$ for any $\omega \neq \omega_0$. Therefore, this fluctuation leads to a value of the action lower than the AP solution, though not necessarily a new minimum.

Similar considerations applied to systems with periodic boundary conditions [23,37–39] lead to a closed expression for the unstable frequency ω_0. Unfortunately, such an expression can hardly be obtained for open systems. But the following general conclusion seems to hold in that case as well; namely, for a finite size L and long time limit $t \to \infty$, the first unstable mode is expected to be the fundamental so that the system is driven through a continuous, second-order-like transition [23].

There is another important unanticipated outcome of our approach using the cumulant generating function. It allows us to prove the yet unresolved question of the stability of driven systems in the presence of an applied field $E \neq 0$, e.g., the weakly asymmetric exclusion process (WASEP) [3]. Starting from the stochastic equation (3), the corresponding Lagrangian rewrites $\mathcal{L}_E = [J + D(\rho)\partial_x\rho - E\sigma(\rho)]^2/2\sigma(\rho)$ instead of Eq. (6). The new time-independent AP Hamilton-Jacobi equations

$$\partial_x(D\partial_x q - E\sigma) - \partial_x(\sigma\partial_x p) = 0,$$
$$-D\partial_{xx}p - E\sigma'\partial_x p - \frac{\sigma'}{2}(\partial_x p)^2 = 0 \quad (19)$$

with the same boundary conditions (14) are now obtained from the modified action $S_E(x,\tau) = (D\partial_x q - E\sigma)\partial_x p - (\sigma/2)(\partial_x p)^2 + (p - \lambda x)\partial_\tau q$ instead of Eq. (12). To study the stability of the AP solution, we again evaluate the variation δS^2_E up to second order of the AP action under the effect of a fluctuation δq of the density and δp of its conjugate momentum. δS^2_E is given by the diagonal quadratic form (15) except for the replacement of $(\partial_x p_0)^2$ by $(\partial_x p_0)^2 + 2E\partial_x p_0$. Therefore, unlike the case $E = 0$, we cannot *a priori* conclude that Eq. (16) is a sufficient condition for the stability of the AP solution. However, it happens that we indeed always have $(\partial_x p_0)^2 + 2E\partial_x p_0 > 0$. This is a consequence of the AP equations (19). Defining $u = \partial_x p_0 + E$ allows us to rewrite the second equation of Eqs. (19) under the form [35]

$$\frac{\partial_x u}{u^2 - E^2} = -\frac{\sigma'}{2D}. \quad (20)$$

Next, we define $h(x) \equiv \int dx[\sigma'(q_0)/2D(q_0)]$ for a known AP density profile $q_0(x)$. An integral of Eq. (20) is implicitly obtained in terms of $h(x)$ as $u = E\coth(Eh(x))$. Therefore, $(\partial_x p_0)^2 + 2E\partial_x p_0 = E^2/\sinh^2(Eh(x)) > 0$ for any E and Eq. (16), $D'\sigma' \geq D\sigma''$, remains a sufficient condition for the stability of the AP solution for $E \neq 0$.

An immediate application of the sufficient condition (16) with an applied field E is to examine the validity of the AP for the yet unsolved WASEP [40,41]. The WASEP dynamics is described by the dynamics of the symmetric exclusion process [42,43]; namely, $D = 1$ and $\sigma = 2\rho(1-\rho)$ with a field E. It is thus clear from Eq. (16) that the AP is valid for the WASEP.

It is nevertheless worth noting that in the case of periodic boundary conditions, Eq. (16) is no longer applicable due to the additional constraint of particle conservation. And indeed for periodic systems, the WASEP was found to be unstable and certain values of the current $J = Q/t$ lead to traveling wave solutions [23].

Another problem where the previous approach proves useful is the Kipnis, Marchioro,Presutti (KMP) model [44], whose MFT dynamics is defined by $D = 1$ and $\sigma = 2\rho^2$. Clearly, the KMP model does not satisfy Eq. (16), thus being nonconclusive about its stability. However, by solving numerically the linear differential system (17), we have been able to obtain conclusive evidence regarding the stability of the KMP model for values of the current J [45,46] significantly higher than previously obtained in the literature. This suggests that the KMP model should also be stable for boundary-driven systems, in agreement with Ref. [47].

In summary, we have presented a new quantitative approach to study the stability of boundary-driven systems out of equilibrium. This approach based on the stochastic MFT provides a necessary and sufficient condition expressed by Eqs. (17) and (18) for the stability of the AP solution. It constitutes a generalization of the Le Chatelier principle. Moreover, in that framework, we have been able to prove the stability of the (boundary-driven) WASEP model.

Finally, we wish to give a glimpse of additional potential applications of the MFT in the realm of quantum or wave

systems. A list of relevant examples includes thermal conductance in quantum chains [10], cold atoms [48], polarized spins injected into superconductors [16], and coherent mesoscopic transport in a random potential [14]. For these out-of-equilibrium systems, the MFT allows us to obtain the full counting statistics, and other stationary properties, e.g., density correlations [49].

For mesoscopic wave transport, a successful approach has been proposed [15] based on a Langevin equation as in Eq. (3) and which relates the current density $j_d(\mathbf{r})$ in the diffusive regime to the local intensity $I(\mathbf{r})$ of the wave, an equivalent of the density ρ in Eq. (3). The corresponding diffusion coefficient is constant and the white noise term is characterized by the function $\sigma(I) \propto I^2$, a result analogous to the aforementioned KMP model. The density correlations obtained from the MFT corroborates this relation between the KMP process [49] and the transport of classical waves through disordered media [14], as well as the symmetric exclusion process [49] and the transport of electrons in a disordered metal [14].

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