

Supporting Information

Hurtado et al. 10.1073/pnas.1013209108

SI Discussion

Hydrodynamic Fluctuation Theory. The evolution of the system of interest is described by the following Langevin equation

$$\partial_t \rho(\mathbf{r}, t) = -\nabla \cdot (\mathbf{Q}_E[\rho(\mathbf{r}, t)] + \boldsymbol{\xi}(\mathbf{r}, t)), \quad [\text{S1}]$$

which expresses the local conservation of certain physical observable. Here $\rho(\mathbf{r}, t)$ is the density field, $\mathbf{j}(\mathbf{r}, t) \equiv \mathbf{Q}_E[\rho(\mathbf{r}, t)] + \boldsymbol{\xi}(\mathbf{r}, t)$ is the fluctuating current, with local average $\mathbf{Q}_E[\rho(\mathbf{r}, t)]$, and $\boldsymbol{\xi}(\mathbf{r}, t)$ is a Gaussian white noise with zero mean and characterized by a variance (or mobility) $\sigma[\rho(\mathbf{r}, t)]$. Notice that the current functional includes in general the effect of a conservative external field, $\mathbf{Q}_E[\rho(\mathbf{r}, t)] = \mathbf{Q}[\rho(\mathbf{r}, t)] + \sigma[\rho(\mathbf{r}, t)]\mathbf{E}$. Using a path integral formulation (1), the probability of observing a given history $\{\rho(\mathbf{r}, t), \mathbf{j}(\mathbf{r}, t)\}_0^\tau$ of duration τ for the density and current fields can be written as

$$P(\{\rho, \mathbf{j}\}_0^\tau) \sim \exp(+L^d I_\tau[\rho, \mathbf{j}]), \quad [\text{S2}]$$

where L is the system linear size, d is the dimensionality, and the functional $I_\tau[\rho, \mathbf{j}]$ is

$$I_\tau[\rho, \mathbf{j}] = -\int_0^\tau dt \int d\mathbf{r} \frac{(\mathbf{j}(\mathbf{r}, t) - \mathbf{Q}_E[\rho(\mathbf{r}, t)])^2}{2\sigma[\rho(\mathbf{r}, t)]}, \quad [\text{S3}]$$

with $\rho(\mathbf{r}, t)$ and $\mathbf{j}(\mathbf{r}, t)$ coupled via the continuity equation

$$\partial_t \rho(\mathbf{r}, t) + \nabla \cdot \mathbf{j}(\mathbf{r}, t) = 0. \quad [\text{S4}]$$

In this way the probability of each history $\{\rho, \mathbf{j}\}_0^\tau$ has a Gaussian weight around the average local behavior given by $\mathbf{Q}_E[\rho(\mathbf{r}, t)]$. Eqs. S2 and S3 are equivalent to the hydrodynamic fluctuation theory recently proposed by Bertini et al. (2–4). The probability $P_\tau(\mathbf{J})$ of observing a space- and time-averaged empirical current \mathbf{J} , defined as

$$\mathbf{J} = \frac{1}{\tau} \int_0^\tau dt \int d\mathbf{r} \mathbf{j}(\mathbf{r}, t), \quad [\text{S5}]$$

can be obtained from the path integral of $P(\{\rho, \mathbf{j}\}_0^\tau)$ restricted to histories $\{\rho, \mathbf{j}\}_0^\tau$ compatible with a given \mathbf{J} ,

$$P_\tau(\mathbf{J}) = \int \mathcal{D}\rho \mathcal{D}\mathbf{j} P(\{\rho, \mathbf{j}\}_0^\tau) \delta\left(\mathbf{J} - \frac{1}{\tau} \int_0^\tau dt \int d\mathbf{r} \mathbf{j}(\mathbf{r}, t)\right). \quad [\text{S6}]$$

This probability scales for long times as $P_\tau(\mathbf{J}) \sim \exp[+\tau L^d G(\mathbf{J})]$, and the current large deviation function (LDF) $G(\mathbf{J})$ can be related to $I_\tau[\rho, \mathbf{j}]$ via a simple saddle-point calculation in the long-time limit,

$$G(\mathbf{J}) = \lim_{\tau \rightarrow \infty} \frac{1}{\tau} \max_{\rho, \mathbf{j}} I_\tau[\rho, \mathbf{j}], \quad [\text{S7}]$$

subject to constraints [S4] and [S5]. The density and current fields solution of this variational problem, denoted here as $\rho_0(\mathbf{r}, t; \mathbf{J})$ and $\mathbf{j}_0(\mathbf{r}, t; \mathbf{J})$, can be interpreted as the optimal path the system follows to sustain a long-time current fluctuation \mathbf{J} . It is worth emphasizing here that the existence of an optimal path rests on the presence of a selection principle at play, namely a long-

time, large-size limit that selects, among all possible paths compatible with a given fluctuation, an optimal one via a saddle-point mechanism. Eq. S7 defines a complex spatiotemporal problem whose solution remains challenging in most cases (1–8). However, the following hypotheses greatly reduce its complexity:

- i. We assume that the optimal profiles responsible of a given current fluctuation are time-independent (7), $\rho_0(\mathbf{r}; \mathbf{J})$ and $\mathbf{j}_0(\mathbf{r}; \mathbf{J})$. This, together with the continuity equation, implies that the optimal current vector field is divergence-free, $\nabla \cdot \mathbf{j}_0(\mathbf{r}; \mathbf{J}) = 0$.
- ii. A further simplification consists in assuming that the optimal current field is in fact constant across space, so $\mathbf{j}_0(\mathbf{r}; \mathbf{J}) = \mathbf{J}$.

Provided that these hypotheses hold, the current LDF can be written as

$$G(\mathbf{J}) = -\min_{\rho(\mathbf{r})} \int \frac{(\mathbf{J} - \mathbf{Q}_E[\rho(\mathbf{r})])^2}{2\sigma[\rho(\mathbf{r})]} d\mathbf{r}. \quad [\text{S8}]$$

The optimal density profile is thus solution of the following differential equation

$$\frac{\delta \omega_2[\rho(\mathbf{r})]}{\delta \rho(\mathbf{r}')} - 2\mathbf{J} \cdot \frac{\delta \omega_1[\rho(\mathbf{r})]}{\delta \rho(\mathbf{r}')} + \mathbf{J}^2 \frac{\delta \omega_0[\rho(\mathbf{r})]}{\delta \rho(\mathbf{r}')} = 0, \quad [\text{S9}]$$

where $\frac{\delta}{\delta \rho(\mathbf{r}')}$ stands for functional derivative, and

$$\omega_n[\rho(\mathbf{r})] \equiv \int d\mathbf{r} \mathbf{W}_n[\rho(\mathbf{r})] \quad \text{with} \quad \mathbf{W}_n[\rho(\mathbf{r})] \equiv \frac{\mathbf{Q}_E^n[\rho(\mathbf{r})]}{\sigma[\rho(\mathbf{r})]}. \quad [\text{S10}]$$

For time-reversible systems, one can see that the evolution operator in the Fokker–Planck formulation of Eq. S1 obeys a local detailed balance condition, and

$$\mathbf{W}_1[\rho(\mathbf{r})] = \frac{\mathbf{Q}_E[\rho(\mathbf{r})]}{\sigma[\rho(\mathbf{r})]} = -\nabla \frac{\delta \mathcal{H}[\rho]}{\delta \rho}, \quad [\text{S11}]$$

where $\mathcal{H}[\rho(\mathbf{r})]$ is the system Hamiltonian. In this case, by using vector integration by parts, it is easy to show that

$$\frac{\delta}{\delta \rho(\mathbf{r}')} \int d\mathbf{r} \mathbf{W}_1[\rho(\mathbf{r})] \cdot \mathcal{A}(\mathbf{r}) = -\frac{\delta}{\delta \rho(\mathbf{r}')} \int_{\partial \Lambda} d\Gamma \frac{\delta \mathcal{H}[\rho]}{\delta \rho} \mathcal{A}(\mathbf{r}) \cdot \hat{n} = 0, \quad [\text{S12}]$$

for any divergence-free vector field $\mathcal{A}(\mathbf{r})$. The second integral is taken over the boundary $\partial \Lambda$ of the domain Λ where the system is defined, and \hat{n} is the unit vector normal to the boundary at each point. In particular, by taking $\mathcal{A}(\mathbf{r}) = \mathbf{J}$ constant, Eq. S12 implies that $\delta \omega_1[\rho(\mathbf{r})]/\delta \rho(\mathbf{r}') = 0$. Hence for time-reversible systems the optimal profile $\rho_0(\mathbf{r}; \mathbf{J})$ remains invariant under rotations of the current \mathbf{J} , see Eq. S9, and this allows us to prove the isometric fluctuation relation (IFR), Eqs. 1 and 8 in the main text.

We can now relax hypothesis (ii) above and study cases where the current profile is not constant. Let $P_\tau[\mathcal{J}(\mathbf{r})]$ be the probability of observing a time-averaged current field $\mathcal{J}(\mathbf{r}) = \tau^{-1} \int_0^\tau dt \mathbf{j}(\mathbf{r}, t)$. Notice that this vector field must be divergence-free because of hypothesis (i). This probability also obeys a large deviation principle, $P_\tau[\mathcal{J}(\mathbf{r})] \sim \exp(+\tau L^d G[\mathcal{J}(\mathbf{r})])$, with a current LDF

equivalent to that in Eq. S8 but with a space-dependent current field $\mathcal{F}(\mathbf{r})$. The optimal density profile $\rho_0[\mathbf{r}; \mathcal{F}(\mathbf{r})]$ is now solution of

$$\frac{\delta}{\delta\rho(\mathbf{r}')} \int d\mathbf{r} (W_2[\rho(\mathbf{r})] - 2\mathcal{F}(\mathbf{r}) \cdot \mathbf{W}_1[\rho(\mathbf{r})] + \mathcal{F}^2(\mathbf{r})W_0[\rho(\mathbf{r})]) = 0, \quad [\text{S13}]$$

which is the equivalent to Eq. S9 in this case. For time-reversible systems condition [S12] holds and $\rho_0[\mathbf{r}; \mathcal{F}(\mathbf{r})]$ remains invariant under (local or global) rotations of $\mathcal{F}(\mathbf{r})$. In this way we can simply relate $P_\tau[\mathcal{F}(\mathbf{r})]$ with the probability of any other divergence-free current field $\mathcal{F}'(\mathbf{r})$ locally isometric to $\mathcal{F}(\mathbf{r})$ (i.e., $\mathcal{F}'(\mathbf{r})^2 = \mathcal{F}(\mathbf{r})^2 \forall \mathbf{r}$) via a generalized isometric fluctuation relation, see Eq. 12 in the paper. Notice that in general an arbitrary local or global rotation of a divergence-free vector field does not conserve the zero-divergence property, so this constraints the current fields and/or local rotations for which this generalized IFR applies.

The large deviation function for the space- and time-averaged current, $G(\mathbf{J})$, can be related to $G[\mathcal{F}(\mathbf{r})]$ via a contraction principle

$$G(\mathbf{J}) = \max_{\substack{\mathcal{F}(\mathbf{r}): \nabla \cdot \mathcal{F}(\mathbf{r})=0 \\ \mathbf{J} = \int d\mathbf{r} \mathcal{F}(\mathbf{r})}} G[\mathcal{F}(\mathbf{r})]. \quad [\text{S14}]$$

The optimal, divergence-free current field $\mathcal{F}_0(\mathbf{r}; \mathbf{J})$ solution of this variational problem may have spatial structure in general. However, numerical results and phenomenological arguments strongly suggest that the constant solution, $\mathcal{F}_0(\mathbf{r}; \mathbf{J}) = \mathbf{J}$, is the optimizer at least for a wide interval of current fluctuations, showing that hypothesis (ii) above is not only plausible but also well justified on physical grounds. In any case, the range of validity of this hypothesis can be explored by studying the limit of local stability of the constant current solution using tools similar to those in ref. 8.

Hypotheses (i) and (ii) are the straightforward generalization to d -dimensional systems of the Additivity Principle recently conjectured by Bodineau and Derrida for one-dimensional diffusive systems (7). This conjecture, which has been recently confirmed for a broad current interval in extensive simulations of a general diffusion model (5, 6), is however known to break down in some special cases for extreme current fluctuations, where time-dependent profiles in the form of traveling waves propagating along the current direction may emerge (2–4, 8). As in previous cases, we can now study the probability $P(\{\mathbf{j}(\mathbf{r}, t)\}_0^\tau)$ of observing a particular history for the current field, which can be written as the path integral of the probability in Eq. S2 over histories of the density field $\{\rho(\mathbf{r}, t)\}_0^\tau$ coupled to the desired current field via the continuity Eq. S4 at every point in space and time. This probability obeys another large deviation principle, with an optimal history of the density field $\{\rho_0(\mathbf{r}, t)\}_0^\tau$ that is solution of an equation similar to Eq. S13 but with time-dependent profiles. However, as opposed to the cases above, the current field $\mathbf{j}(\mathbf{r}, t)$ is not necessarily divergence-free because of the time-dependence of the associated $\rho_0(\mathbf{r}, t)$, resulting in a violation of condition [S12]. In this way the optimal $\rho_0(\mathbf{r}, t)$ depends on both $\mathbf{j}(\mathbf{r}, t)$ and $\mathbf{j}(\mathbf{r}, t)^2$ so it does not remain invariant under (local or global) instantaneous rotations of the current field, resulting in a violation of the generalized isometric fluctuation relation in the time-dependent regime.

Notice that the dynamic phase transition to time-dependent optimal paths is expected to occur only for extreme current fluctuations, thus rendering valid the isometric fluctuation relations for a wide, subcritical current interval. Interestingly, we can use

the IFR to detect such dynamic phase transition. If we measure $P_\tau[\mathcal{F}(\mathbf{r})]$ in a system described by Eq. S1 at the macroscopic level, finding that the measured probabilities do not obey the generalized IFR, then we can conclude that such a violation of IFR is due to the onset of time-dependent optimal profiles, thus signaling the dynamic phase transition. On the other hand, breakdown of the standard IFR (for space- and time-averaged currents) may signal the onset of space-dependent, divergence-free optimal current profiles or the aforementioned dynamic phase transition. In this way, the combined use of the IFR and its generalizations is capable of a full characterization of the instabilities that characterize the fluctuating behavior of the system at hand (8).

Constants of Motion. A sufficient condition for the IFR to hold is that

$$\frac{\delta\omega_1[\rho(\mathbf{r})]}{\delta\rho(\mathbf{r}')} = 0, \quad [\text{S15}]$$

with the functional $\omega_1[\rho(\mathbf{r})]$ defined in Eq. S10 above. We have shown that condition [S15] follows from the time-reversibility of the dynamics, in the sense that the evolution operator in the Fokker–Planck formulation of Eq. S1 obeys a local detailed balance condition, see Eq. S12. Condition [S15] implies that $\omega_1[\rho(\mathbf{r})]$ is in fact a constant of motion, ϵ , independent of the profile $\rho(\mathbf{r})$. Therefore we can use an arbitrary profile $\rho(\mathbf{r})$, compatible with boundary conditions, to compute ϵ . We now choose boundary conditions to be gradient-like in the \hat{x} -direction, with densities ρ_L and ρ_R at the left and right reservoirs, respectively, and periodic boundary conditions in all other directions. Given these boundaries, we now select a linear profile

$$\rho(\mathbf{r}) = \rho_L + (\rho_R - \rho_L)x, \quad [\text{S16}]$$

to compute ϵ , with $x \in [0, 1]$, and assume very general forms for the current and mobility functionals

$$\mathbf{Q}[\rho(\mathbf{r})] \equiv D_{0,0}[\rho] \nabla \rho + \sum_{n,m>0} D_{nm}[\rho] (\nabla^m \rho)^{2n} \nabla \rho,$$

$$\sigma[\rho(\mathbf{r})] \equiv \sigma_{0,0}[\rho] + \sum_{n,m>0} \sigma_{nm}[\rho] (\nabla^m \rho)^{2n},$$

whereas a convention we denote as $F[\rho]$ a generic functional of the profile but not of its derivatives. It is now easy to show that $\epsilon = \epsilon \hat{x} + \mathbf{E}$, with

$$\epsilon = \int_{\rho_L}^{\rho_R} d\rho \frac{D_{0,0}(\rho) + \sum_{n>0} D_{n1}(\rho) (\rho_R - \rho_L)^{2n}}{\sigma_{0,0}(\rho) + \sum_{m>0} \sigma_{m1}(\rho) (\rho_R - \rho_L)^{2m}}, \quad [\text{S17}]$$

and \hat{x} the unit vector along the gradient direction. In a similar way, if the following condition holds

$$\frac{\delta}{\delta\rho(\mathbf{r}')} \int \mathbf{Q}[\rho(\mathbf{r})] d\mathbf{r} = 0, \quad [\text{S18}]$$

together with time-reversibility, Eq. S15, the system can be shown to obey an extended isometric fluctuation relation that links any current fluctuation \mathbf{J} in the presence of an external field \mathbf{E} with any other isometric current fluctuation \mathbf{J}' in the presence of an arbitrarily rotated external field \mathbf{E}^* , and reduces to the standard IFR for $\mathbf{E} = \mathbf{E}^*$, see Eq. 11 in the paper. Condition [S18] implies that $\nu \equiv \int \mathbf{Q}[\rho(\mathbf{r})] d\mathbf{r}$ is another constant of motion, which can be now written as $\nu = \nu \hat{x}$, with

$$\nu = \int_{\rho_L}^{\rho_R} d\rho \left[D_{0,0}(\rho) + \sum_{n>0} D_{n1}(\rho) (\rho_R - \rho_L)^{2n} \right]. \quad [\text{S19}]$$

$$\varepsilon = \int_{\rho_R}^{\rho_L} \frac{D(\rho)}{\sigma(\rho)} d\rho, \quad \nu = \int_{\rho_R}^{\rho_L} D(\rho) d\rho,$$

As an example, for a diffusive system $\mathbf{Q}[\rho(\mathbf{r})] = -D[\rho]\nabla\rho(\mathbf{r})$, with $D[\rho]$ the diffusivity functional, and the above equations yield the familiar results

for a standard local mobility $\sigma[\rho]$.

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