

Symmetries in fluctuations far from equilibrium

Pablo I. Hurtado¹, Carlos Pérez-Espigares, Jesús J. del Pozo, and Pedro L. Garrido

Departamento de Electromagnetismo y Física de la Materia, and Instituto Carlos I de Física Teórica y Computacional, Universidad de Granada, Granada 18071, Spain

Edited by Joel L. Lebowitz, Center for Mathematical Sciences Research, Piscataway, NJ, and approved February 22, 2011 (received for review September 6, 2010)

Fluctuations arise universally in nature as a reflection of the discrete microscopic world at the macroscopic level. Despite their apparent noisy origin, fluctuations encode fundamental aspects of the physics of the system at hand, crucial to understand irreversibility and nonequilibrium behavior. To sustain a given fluctuation, a system traverses a precise optimal path in phase space. Here we show that by demanding invariance of optimal paths under symmetry transformations, new and general fluctuation relations valid arbitrarily far from equilibrium are unveiled. This opens an unexplored route toward a deeper understanding of nonequilibrium physics by bringing symmetry principles to the realm of fluctuations. We illustrate this concept studying symmetries of the current distribution out of equilibrium. In particular we derive an isometric fluctuation relation that links in a strikingly simple manner the probabilities of any pair of isometric current fluctuations. This relation, which results from the time-reversibility of the dynamics, includes as a particular instance the Gallavotti–Cohen fluctuation theorem in this context but adds a completely new perspective on the high level of symmetry imposed by time-reversibility on the statistics of nonequilibrium fluctuations. The new symmetry implies remarkable hierarchies of equations for the current cumulants and the nonlinear response coefficients, going far beyond Onsager's reciprocity relations and Green–Kubo formulas. We confirm the validity of the new symmetry relation in extensive numerical simulations, and suggest that the idea of symmetry in fluctuations as invariance of optimal paths has far-reaching consequences in diverse fields.

large deviations | rare events | hydrodynamics | transport | entropy production

Large fluctuations, though rare, play an important role in many fields of science as they crucially determine the fate of a system (1). Examples range from chemical reaction kinetics or the escape of metastable electrons in nanoelectronic devices to conformational changes in proteins, mutations in DNA, and nucleation events in the primordial universe. Remarkably, the statistics of these large fluctuations contains deep information on the physics of the system of interest (2, 3). This is particularly important for systems far from equilibrium, where no general theory exists up to date capable of predicting macroscopic and fluctuating behavior in terms of microscopic physics, in a way similar to equilibrium statistical physics. The consensus is that the study of fluctuations out of equilibrium may open the door to such general theory. As most nonequilibrium systems are characterized by currents of locally conserved observables, understanding current statistics in terms of microscopic dynamics has become one of the main objectives of nonequilibrium statistical physics (2–17). Pursuing this line of research is both of fundamental as well as practical importance. At the theoretical level, the function controlling current fluctuations can be identified as the nonequilibrium analog of the free-energy functional in equilibrium systems (2–5), from which macroscopic properties of a nonequilibrium system can be obtained (including its most prominent features, as for instance the ubiquitous long range correlations (18, 19), etc.) On the other hand, the physics of most modern mesoscopic devices is characterized by large fluctuations that de-

termine their behavior and function. In this way understanding current statistics in these systems is of great practical significance.

Despite the considerable interest and efforts on these issues, exact and general results valid arbitrarily far from equilibrium are still very scarce. The reason is that, whereas in equilibrium phenomena dynamics is irrelevant and the Gibbs distribution provides all the necessary information, in nonequilibrium physics dynamics plays a dominant role, even in the simplest situation of a nonequilibrium steady state (2–5). However, there is a remarkable exception to this absence of general results that has triggered an important surge in activity since its formulation in the mid nineties. This is the fluctuation theorem, first discussed in the context of simulations of sheared fluids (14), and formulated rigorously by Gallavotti and Cohen under very general assumptions (15). This theorem, which implies a relation between the probabilities of a given current fluctuation and the inverse event, is a deep statement on the subtle consequences of time-reversal symmetry of microscopic dynamics at the macroscopic, irreversible level. Particularly important here is the observation that symmetries are reflected at the fluctuating macroscopic level arbitrarily far from equilibrium. Inspired by this illuminating result, we explore in this paper the behavior of the current distribution under symmetry transformations (20). Key to our analysis is the observation that, to facilitate a given current fluctuation, the system traverses a well-defined optimal path in phase space (2–8, 21). This path is, under very general conditions, invariant under certain symmetry transformations on the current. Using this invariance we show that for d -dimensional, time-reversible systems described by a locally conserved field and possibly subject to a boundary-induced gradient and an external field \mathbf{E} , the probability $P_\tau(\mathbf{J})$ of observing a current \mathbf{J} averaged over a long time τ obeys an isometric fluctuation relation (IFR)

$$\lim_{\tau \rightarrow \infty} \frac{1}{\tau} \ln \left[\frac{P_\tau(\mathbf{J})}{P_\tau(\mathbf{J}')} \right] = \boldsymbol{\epsilon} \cdot (\mathbf{J} - \mathbf{J}'), \quad [1]$$

for any pair of isometric current vectors, $|\mathbf{J}| = |\mathbf{J}'|$. Here $\boldsymbol{\epsilon} = \boldsymbol{\epsilon} + \mathbf{E}$ is a constant vector directly related to the rate of entropy production in the system, which depends on the boundary baths via $\boldsymbol{\epsilon}$ (see below).

The above equation, which includes as a particular case the Gallavotti–Cohen (GC) result for $\mathbf{J}' = -\mathbf{J}$, relates in a strikingly simple manner the probability of a given fluctuation \mathbf{J} with the likelihood of any other current fluctuation on the d -dimensional hypersphere of radius $|\mathbf{J}|$, see Fig. 1, projecting a complex d -dimensional problem onto a much simpler one-dimensional theory. Unlike the GC relation, which is a nondifferentiable symmetry involving the inversion of the current sign, $\mathbf{J} \rightarrow -\mathbf{J}$, Eq. 1 is valid for arbitrary changes in orientation of the current vector. This

Author contributions: P.I.H. and P.L.G. designed research; P.I.H., C.P.-E., J.J.d.P., and P.L.G. performed research; P.I.H., C.P.-E., J.J.d.P., and P.L.G. analyzed data; and P.I.H. wrote the paper.

The authors declare no conflict of interest.

This article is a PNAS Direct Submission.

¹To whom correspondence should be addressed. E-mail: phurtado@onsager.ugr.es.

This article contains supporting information online at www.pnas.org/lookup/suppl/doi:10.1073/pnas.1013209108/-DCSupplemental.

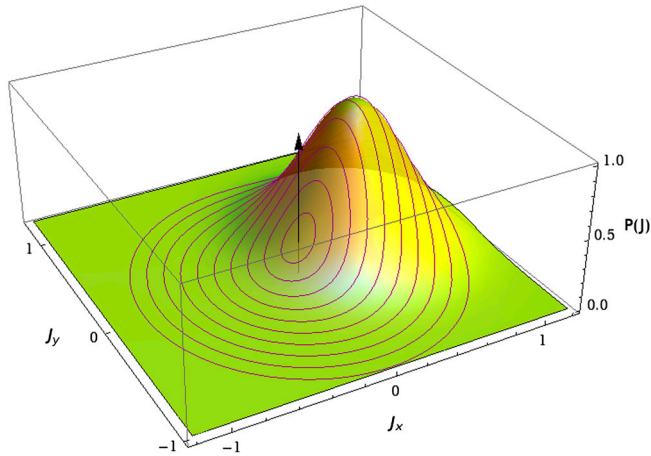


Fig. 1. The isometric fluctuation relation at a glance. Sketch of the current distribution in two dimensions, peaked around its average $\langle \mathbf{J} \rangle$, and isometric contour lines for different $|\mathbf{J}|$ s. The isometric fluctuation relation, Eq. 1, establishes a simple relation for the probability of current fluctuations along each of these contour lines.

makes the experimental test of the above relation a feasible problem, as data for current fluctuations involving different orientations around the average can be gathered with enough statistics to ensure experimental accuracy. It is also important to notice that the isometric fluctuation relation is valid for arbitrarily large fluctuations; i.e., even for the non-Gaussian far tails of current distribution. We confirm here the validity of this symmetry in extensive numerical simulations of two different nonequilibrium systems: (i) A simple and very general lattice model of energy diffusion (7, 8, 22), and (ii) a hard-disk fluid in a temperature gradient (23).

Results

The Isometric Fluctuation Relation Our starting point is a continuity equation that describes the macroscopic evolution of a wide class of systems characterized by a locally conserved magnitude (e.g., energy, particle density, momentum, etc.)

$$\partial_t \rho(\mathbf{r}, t) = -\nabla \cdot (\mathbf{Q}_E[\rho(\mathbf{r}, t)] + \xi(\mathbf{r}, t)). \quad [2]$$

Here $\rho(\mathbf{r}, t)$ is the density field, $\mathbf{j}(\mathbf{r}, t) \equiv \mathbf{Q}_E[\rho(\mathbf{r}, t)] + \xi(\mathbf{r}, t)$ is the fluctuating current, with local average $\mathbf{Q}_E[\rho(\mathbf{r}, t)]$, and $\xi(\mathbf{r}, t)$ is a Gaussian white noise characterized by a variance (or mobility) $\sigma[\rho(\mathbf{r}, t)]$. This (conserved) noise term accounts for microscopic random fluctuations at the macroscopic level. Notice that the current functional includes in general the effect of a conservative external field, $\mathbf{Q}_E[\rho(\mathbf{r}, t)] = \mathbf{Q}[\rho(\mathbf{r}, t)] + \sigma[\rho(\mathbf{r}, t)]\mathbf{E}$. Examples of systems described by Eq. 2 range from diffusive systems (2–9), where $\mathbf{Q}[\rho(\mathbf{r}, t)]$ is given by Fourier's (or equivalently Fick's) law, $\mathbf{Q}[\rho(\mathbf{r}, t)] = -D[\rho]\nabla\rho(\mathbf{r}, t)$, to most interacting-particle fluids (24, 25), characterized by a Ginzburg–Landau-type theory for the locally conserved particle density. To completely define the problem, the above evolution equation must be supplemented with appropriate boundary conditions, which may include an external gradient.

We are interested in the probability $P_\tau(\mathbf{J})$ of observing a space- and time-averaged empirical current \mathbf{J} , defined as

$$\mathbf{J} = \frac{1}{\tau} \int_0^\tau dt \int d\mathbf{r} \mathbf{j}(\mathbf{r}, t). \quad [3]$$

This probability obeys a large-deviation principle for long times (26, 27), $P_\tau(\mathbf{J}) \sim \exp[\pm\tau L^d G(\mathbf{J})]$, where L is the system linear size and $G(\mathbf{J}) \leq 0$ is the current large-deviation function (LDF), meaning that current fluctuations away from the average are

exponentially unlikely in time. According to hydrodynamic fluctuation theory (2, 4–6),

$$G(\mathbf{J}) = -\min_{\rho(\mathbf{r})} \int \frac{(\mathbf{J} - \mathbf{Q}_E[\rho(\mathbf{r})])^2}{2\sigma[\rho(\mathbf{r})]} d\mathbf{r}, \quad [4]$$

which expresses the locally-Gaussian nature of fluctuations (6–8). The optimal profile $\rho_0(\mathbf{r}; \mathbf{J})$ solution of the above variational problem can be interpreted as the density profile the system adopts to facilitate a current fluctuation \mathbf{J} (7, 8, 21). To derive Eq. 4 we assumed that (i) the optimal profiles associated to a given current fluctuation are time-independent (2–9, 21), and (ii) the optimal current field has no spatial structure (*SI Text*). This last hypothesis, which greatly simplifies the calculation of current statistics, can be however relaxed for our purposes (as shown below). The probability $P_\tau(\mathbf{J})$ is thus simply the Gaussian weight associated to the optimal profile. Note however that the minimization procedure gives rise to a nonlinear problem that results in general in a current distribution with non-Gaussian tails (2–8).

The optimal profile is solution of the following equation

$$\frac{\delta\omega_2[\rho(\mathbf{r})]}{\delta\rho(\mathbf{r}')} - 2\mathbf{J} \cdot \frac{\delta\omega_1[\rho(\mathbf{r})]}{\delta\rho(\mathbf{r}')} + \mathbf{J}^2 \frac{\delta\omega_0[\rho(\mathbf{r})]}{\delta\rho(\mathbf{r}')} = 0, \quad [5]$$

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

$$\omega_n[\rho(\mathbf{r})] \equiv \int d\mathbf{r} \frac{\mathbf{Q}_E^n[\rho(\mathbf{r})]}{\sigma[\rho(\mathbf{r})]}. \quad [6]$$

Remarkably, the optimal profile $\rho_0(\mathbf{r}; \mathbf{J})$ solution of Eq. 5 depends exclusively on \mathbf{J} and \mathbf{J}^2 . Such a simple quadratic dependence, inherited from the locally Gaussian nature of fluctuations, has important consequences at the level of symmetries of the current distribution. In fact, it is clear from Eq. 5 that the condition

$$\frac{\delta\omega_1[\rho(\mathbf{r})]}{\delta\rho(\mathbf{r}')} = 0, \quad [7]$$

implies that $\rho_0(\mathbf{r}; \mathbf{J})$ will depend exclusively on the magnitude of the current vector, via \mathbf{J}^2 , not on its orientation. In this way, all isometric current fluctuations characterized by a constant $|\mathbf{J}|$ will have the same associated optimal profile, $\rho_0(\mathbf{r}; \mathbf{J}) = \rho_0(\mathbf{r}; |\mathbf{J}|)$, independently of whether the current vector \mathbf{J} points along the gradient direction, against it, or along any arbitrary direction. In other words, the optimal profile is invariant under current rotations if Eq. 7 holds. It turns out that condition [7] follows from the time-reversibility of the dynamics, in the sense that the evolution operator in the Fokker–Planck formulation of Eq. 2 obeys a local detailed balance condition (16, 17). In this case $\mathbf{Q}_E[\rho(\mathbf{r})]/\sigma[\rho(\mathbf{r})] = -\nabla\delta\mathcal{H}[\rho]/\delta\rho$, with $\mathcal{H}[\rho(\mathbf{r})]$ the system Hamiltonian, and condition [7] holds. The invariance of the optimal profile can be now used in Eq. 4 to relate in a simple way the current LDF of any pair of isometric current fluctuations \mathbf{J} and \mathbf{J}' , with $|\mathbf{J}| = |\mathbf{J}'|$,

$$G(\mathbf{J}) - G(\mathbf{J}') = |\epsilon||\mathbf{J}|(\cos\theta - \cos\theta'), \quad [8]$$

where θ and θ' are the angles formed by vectors \mathbf{J} and \mathbf{J}' , respectively, with a constant vector $\epsilon = \epsilon + \mathbf{E}$; see below. Eq. 8 is just an alternative formulation of the isometric fluctuation relation [1]. By letting \mathbf{J} and \mathbf{J}' differ by an infinitesimal angle, the IFR can be cast in a simple differential form, $\partial_\theta G(\mathbf{J}) = |\epsilon||\mathbf{J}| \sin\theta$, which reflects the high level of symmetry imposed by time-reversibility on the current distribution.

The condition $\delta\omega_1[\rho(\mathbf{r})]/\delta\rho(\mathbf{r}') = 0$ can be seen as a conservation law. It implies that the observable $\omega_1[\rho(\mathbf{r})]$ is in fact a constant of motion, $\epsilon \equiv \omega_1[\rho(\mathbf{r})]$, independent of the profile

$\rho(\mathbf{r})$, which can be related with the rate of entropy production via the Gallavotti–Cohen theorem (15–17). In a way similar to Noether’s theorem, the conservation law for ϵ implies a symmetry for the optimal profiles under rotations of the current and a fluctuation relation for the current LDF. This constant can be easily computed under very general assumptions (*SI Text*).

Implications and Generalizations The isometric fluctuation relation, Eq. 1, has far-reaching and nontrivial consequences. First, the IFR implies a remarkable hierarchy of equations for the cumulants of the current distribution, see Eq. 13 in *Materials and Methods*. This hierarchy can be derived starting from the Legendre transform of the current LDF, $\mu(\lambda) = \max_{\mathbf{J}}[G(\mathbf{J}) + \lambda \cdot \mathbf{J}]$, from which all cumulants can be obtained (3), and writing the IFR for $\mu(\lambda)$ in the limit of infinitesimal rotations. As an example, the cumulant hierarchy in two dimensions implies the following relations

$$\begin{aligned} \langle J_x \rangle_\epsilon &= \tau L^2 [\epsilon_x \langle \Delta J_y^2 \rangle_\epsilon - \epsilon_y \langle \Delta J_x \Delta J_y \rangle_\epsilon] \\ \langle J_y \rangle_\epsilon &= \tau L^2 [\epsilon_y \langle \Delta J_x^2 \rangle_\epsilon - \epsilon_x \langle \Delta J_x \Delta J_y \rangle_\epsilon] \end{aligned} \quad [9]$$

$$\begin{aligned} 2\langle \Delta J_x \Delta J_y \rangle_\epsilon &= \tau L^2 [\epsilon_y \langle \Delta J_x^3 \rangle_\epsilon - \epsilon_x \langle \Delta J_x^2 \Delta J_y \rangle_\epsilon] \\ &= \tau L^2 [\epsilon_x \langle \Delta J_y^3 \rangle_\epsilon - \epsilon_y \langle \Delta J_x \Delta J_y^2 \rangle_\epsilon] \\ \langle \Delta J_x^2 \rangle_\epsilon - \langle \Delta J_y^2 \rangle_\epsilon &= \tau L^2 [\epsilon_x \langle \Delta J_x \Delta J_y^2 \rangle_\epsilon - \epsilon_y \langle \Delta J_x^2 \Delta J_y \rangle_\epsilon], \end{aligned} \quad [10]$$

for the first cumulants, with $\Delta J_\alpha \equiv J_\alpha - \langle J_\alpha \rangle_\epsilon$. It is worth stressing that the cumulant hierarchy is valid arbitrarily far from equilibrium. In a similar way, the IFR implies a set of hierarchies for the nonlinear response coefficients, see Eqs. 15–17 in *Materials and Methods*. In our two-dimensional example, let $\chi_{(n_x, n_y)}^{(k_x, k_y)}$ be the response coefficient of the cumulant $\langle \Delta J_x^{n_x} \Delta J_y^{n_y} \rangle_\epsilon$ to order $\epsilon_x^{k_x} \epsilon_y^{k_y}$, with $n = n_x + n_y$ and $k = k_x + k_y$. To the lowest order these hierarchies imply Onsager’s reciprocity symmetries and Green–Kubo relations for the linear response coefficients of the current. They further predict that in fact the linear response matrix is proportional to the identity, so $\chi_{(1,0)}^{(1,0)} = \chi_{(1,0)}^{(0,1)} = \chi_{(2,0)}^{(0,0)} = \chi_{(2,0)}^{(0,0)}$ whereas $\chi_{(1,0)}^{(0,1)} = 0 = \chi_{(1,0)}^{(1,0)}$. The first nonlinear coefficients of the current can be simply written in terms of the linear coefficients of the second cumulants as $\chi_{(1,0)}^{(2,0)} = 2\chi_{(2,0)}^{(1,0)}$ and $\chi_{(1,0)}^{(0,2)} = -2\chi_{(1,1)}^{(1,0)}$, whereas the cross-coefficient reads $\chi_{(1,0)}^{(1,1)} = 2[\chi_{(2,0)}^{(0,1)} + \chi_{(2,0)}^{(0,1)}]$ (symmetric results hold for $n_x = 0, n_y = 1$). Linear response coefficients for the second-order cumulants also obey simple relations (e.g., $\chi_{(2,1)}^{(1,0)} = -\chi_{(2,1)}^{(0,1)}$) and $\chi_{(2,0)}^{(1,0)} + \chi_{(2,0)}^{(0,1)} = \chi_{(2,0,2)}^{(1,0)} + \chi_{(2,0,2)}^{(0,1)}$) and the set of relations continues to arbitrary high orders. In this way hierarchies [15–17], which derive from microreversibility as reflected in the IFR, provide deep insights into nonlinear response theory for nonequilibrium systems (28).

The IFR and the above hierarchies all follow from the invariance of optimal profiles under certain transformations. This idea can be further exploited in more general settings. In fact, by writing explicitly the dependence on the external field \mathbf{E} in Eq. 5 for the optimal profile, one realizes that if $\frac{\delta}{\delta \rho(\mathbf{r})} \int \mathbf{Q}[\rho(\mathbf{r})] d\mathbf{r} = 0$, together with the time-reversibility condition, Eq. 7, the resulting optimal profiles are invariant under independent rotations of the current and the external field. It thus follows that the current LDFs for pairs (\mathbf{J}, \mathbf{E}) and $(\mathbf{J}' = \mathcal{R}\mathbf{J}, \mathbf{E}' = \mathcal{S}\mathbf{E})$, with \mathcal{R}, \mathcal{S} independent rotations, obey a generalized isometric fluctuation relation

$$G_{\mathbf{E}}(\mathbf{J}) - G_{\mathbf{E}'}(\mathbf{J}') = \boldsymbol{\epsilon} \cdot (\mathbf{J} - \mathbf{J}') - \boldsymbol{\nu} \cdot (\mathbf{E} - \mathbf{E}') + \mathbf{J} \cdot \mathbf{E} - \mathbf{J}' \cdot \mathbf{E}', \quad [11]$$

where we write explicitly the dependence of the current LDF on the external field. The vector $\boldsymbol{\nu} \equiv \int \mathbf{Q}[\rho(\mathbf{r})] d\mathbf{r}$ is now another constant of motion, independent of $\rho(\mathbf{r})$, which can be easily computed (*SI Text*). For a fixed boundary gradient, the above equation relates 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 $\mathbf{E} = \mathbf{E}'$. Condition $\frac{\delta}{\delta \rho(\mathbf{r})} \int \mathbf{Q}[\rho(\mathbf{r})] d\mathbf{r} = 0$ is rather general, as most time-reversible systems with a local mobility $\sigma[\rho]$ do fulfill this condition (e.g., diffusive systems).

The IFR can be further generalized to cases where the current profile is not constant, relaxing hypothesis (ii) above. Let $P_\tau[\mathcal{F}(\mathbf{r})]$ be the probability of observing a time-averaged current field $\mathcal{F}(\mathbf{r}) = \tau^{-1} \int_0^\tau dt \mathbf{j}(\mathbf{r}, t)$. This vector field must have zero divergence because it is coupled via the continuity equation to an optimal density profile that is assumed to be time-independent; see *SI Text* and hypothesis (i) above. Because of time-reversibility, $\mathbf{Q}_{\mathbf{E}}[\rho(\mathbf{r})]/\sigma[\rho(\mathbf{r})] = -\nabla \delta \mathcal{H}[\rho]/\delta \rho$ and it is easy to show in the equation for the optimal density profile that the term linear in $\mathcal{F}(\mathbf{r})$ vanishes, so $\rho_0[\mathbf{r}; \mathcal{F}(\mathbf{r})]$ remains invariant under (local or global) rotations of $\mathcal{F}(\mathbf{r})$; see *SI Text*. In this way, for any divergence-free current field $\mathcal{F}'(\mathbf{r})$ locally isometric to $\mathcal{F}(\mathbf{r})$, so $\mathcal{F}'(\mathbf{r})^2 = \mathcal{F}(\mathbf{r})^2 \forall \mathbf{r}$, we can write a generalized isometric fluctuation relation

$$\lim_{\tau \rightarrow \infty} \frac{1}{\tau} \ln \left[\frac{P_\tau[\mathcal{F}(\mathbf{r})]}{P_\tau[\mathcal{F}'(\mathbf{r})]} \right] = \int_{\partial \Lambda} d\Gamma \frac{\delta \mathcal{H}[\rho]}{\delta \rho} \hat{n} \cdot [\mathcal{F}'(\mathbf{r}) - \mathcal{F}(\mathbf{r})], \quad [12]$$

where the integral (whose result is independent of $\rho(\mathbf{r})$) 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. Eq. 12 generalizes the IFR to situations where hypothesis (ii) is violated, opening the door to isometries based on local (in addition to global) rotations. As a corollary, we show in *SI Text* that a similar generalization of the isometric fluctuation symmetry does not exist whenever optimal profiles become time-dependent, so the IFR breaks down in the regime where hypothesis (i) is violated. In this way, we may use violations of the IFR and its generalizations to detect the instabilities that characterize the fluctuating behavior of the system at hand (2, 9).

Checking the Isometric Fluctuation Relation We have tested the validity of the IFR in extensive numerical simulations of two different nonequilibrium systems. The first one is a simple and very general model of energy diffusion (7, 8, 22) defined on a two-dimensional (2D) square lattice with L^2 sites. Each site is characterized by an energy e_i , $i \in [1, L^2]$, and models a harmonic oscillator that is mechanically uncoupled from its nearest neighbors but interact with them via a stochastic energy-redistribution process. Dynamics thus proceeds through random energy exchanges between randomly chosen nearest neighbors. In addition, left and right boundary sites may interchange energy with boundary baths at temperatures T_L and T_R , respectively, while periodic boundary conditions hold in the vertical direction. For $T_L \neq T_R$ the systems reaches a nonequilibrium steady-state characterized, in the absence of external field (the case studied here), by a linear energy profile $\rho_{st}(\mathbf{r}) = T_L + x(T_R - T_L)$ and a nonzero average current given by Fourier’s law. This model plays a fundamental role in nonequilibrium statistical physics as a testbed to assess new theoretical advances, and represents at a coarse-grained level a large class of diffusive systems of technological and theoretical interest (7, 8). The model is described at the macroscopic level by Eq. 2 with a diffusive current term $\mathbf{Q}[\rho(\mathbf{r}, t)] = -D[\rho] \nabla \rho$ with $D[\rho] = \frac{1}{2}$ and $\sigma[\rho] = \rho^2$, and it turns out to be an optimal candidate to test

that act as deterministic heat baths, see Fig. 4, *Bottom Inset*. This is achieved by keeping constant the total kinetic energy within each lateral band via a global, instantaneous rescaling of the velocity of bath particles after bath-bulk particle collisions. This heat bath mechanism has been shown to efficiently thermostat the fluid (23), and has the important advantage of being deterministic. As for the previous diffusive model, we performed a large number of steady-state simulations of long duration ($\tau > 2N$ collisions per particle) for $T_L = 4$ and $T_R = 1$, accumulating statistics for the current \mathbf{J} and measuring the average temperature profile associated to each \mathbf{J} . Fig. 4 shows the linear collapse of $|\mathbf{J}|^{-1}[G(\mathbf{J}) - G(\mathbf{J}')] - G(\mathbf{J}')$ as a function of $\cos\theta - \cos\theta'$ for different values of $|\mathbf{J}|$, confirming the validity of the IFR for this hard-disk fluid in the moderate range of current fluctuations that we could access. Moreover, the measured optimal profiles for different isometric current fluctuations all nicely collapse onto single curves, see Fig. 4, *Top Inset*, confirming their rotational invariance.

It is interesting to notice that the hard-disk fluid is a fully hydrodynamic system, with 4 different locally conserved coupled fields possibly subject to memory effects, defining a far more complex situation than the one studied here, see Eq. 2. Therefore the validity of IFR in this context suggests that this fluctuation relation, based on the invariance of optimal profiles under symmetry transformations, is in fact a rather general result valid for arbitrary fluctuating hydrodynamic systems.

A few remarks are now in order. First, as a corollary to the IFR, it should be noted that for time-reversible systems with additive fluctuations (i.e., with a constant, profile-independent mobility σ) the optimal profile associated to a given current fluctuation is in fact independent of \mathbf{J} , see Eq. 5, and hence equal to the stationary profile. In this case it is easy to show that current fluctuations are Gaussian, with $G(\mathbf{J}) = \epsilon \cdot (\mathbf{J} - \langle \mathbf{J} \rangle_\epsilon) + \sigma^{-1}(\mathbf{J}^2 - \langle \mathbf{J} \rangle_\epsilon^2)$. This is the case, for instance, of model B in the Hohenberg–Halperin classification (25)*. On the other hand, it should be noticed that the time-reversibility condition for the IFR to hold, Eq. 7, is just a sufficient but not necessary condition. In fact, we cannot discard the possibility of time-irreversible systems such that, only for the optimal profiles, $\delta\omega_1[\rho(\mathbf{r})]/\delta\rho(\mathbf{r}')|_{\rho_0} = 0$.

Discussion

The IFR is a consequence of time-reversibility for systems in the hydrodynamic scaling limit, and reveals an unexpected high level of symmetry in the statistics of nonequilibrium fluctuations. It generalizes and comprises the Gallavotti–Cohen fluctuation theorem for currents, relating the probabilities of an event not only with its time-reversal but with any other isometric fluctuation. This has important consequences in the form of hierarchies for the current cumulants and the linear and nonlinear response coefficients, which hold arbitrarily far from equilibrium and can be readily tested in experiments. A natural question thus concerns the level of generality of the isometric fluctuation relation. In this paper we have demonstrated the IFR for a broad class of systems characterized at the macroscale by a single conserved field, using the tools of hydrodynamic fluctuation theory (HFT). This theoretical framework, summarized in the path large-deviation functional, Eq. S3, has been rigorously proven for a number of interacting-particle systems (2–5), but it is believed to remain valid for a much larger class of systems. The key is that the Gaussian nature of local fluctuations, which lies at the heart of the approach, is expected to emerge for most situations in the appropriate macroscopic limit as a result of a central limit theorem: Although microscopic interactions can be extremely complicated,

the ensuing fluctuations of the slow hydrodynamic fields result from the sum of an enormous amount of random events at the microscale that give rise to Gaussian statistics. There exist of course anomalous systems for which local fluctuations at the macroscale can be non-Gaussian. In these cases we cannot discard that a modified version of the IFR could remain valid, though the analysis would be certainly more complicated. Furthermore, our numerical results show that the IFR remains true even in cases where it is not clear whether the HFT applies, strongly supporting the validity of this symmetry for arbitrary fluctuating hydrodynamic systems.

A related question is the demonstration of the IFR starting from microscopic dynamics. Techniques similar to those in refs. 16 and 31, which derive the Gallavotti–Cohen fluctuation theorem from the spectral properties of the microscopic stochastic evolution operator, can prove useful for this task. However, to prove the IFR these techniques must be supplemented with additional insights on the asymptotic properties of the microscopic transition rates as the macroscopic limit is approached. In this way we expect finite-size corrections to the IFR that decay with the system size, as it is in fact observed in our simulations for the energy diffusion model, see Fig. 3. Also interesting is the possibility of an IFR for discrete isometries related with the underlying lattice in stochastic models. These open questions call for further study.

We have shown in this paper how symmetry principles come forth in fluctuations far from equilibrium. By demanding invariance of the optimal path responsible of a given fluctuation under symmetry transformations, we unveiled a remarkable and very general isometric fluctuation relation for time-reversible systems that relates in a simple manner the probability of any pair of isometric current fluctuations. Invariance principles of this kind can be applied with great generality in diverse fields where fluctuations play a fundamental role, opening the door to further exact and general results valid arbitrarily far from equilibrium. This is particularly relevant in mesoscopic biophysical systems, where relations similar to the isometric fluctuation relation might be used to efficiently measure free-energy differences in terms of work distributions (32). Other interesting issues concern the study of general fluctuation relations emerging from the invariance of optimal paths in full hydrodynamical systems with several conserved fields, or the quantum analog of the isometric fluctuation relation in full counting statistics.

Materials and Methods

We now exploit the IFR (1) to derive a set of hierarchies for the current cumulants and the linear and nonlinear response coefficients. The moment-generating function associated to $P_\epsilon(\mathbf{J})$, defined as $\Pi_\epsilon(\lambda) = \int P_\epsilon(\mathbf{J}) \exp(\tau L^d \lambda \cdot \mathbf{J}) d\mathbf{J}$, scales for long times as $\Pi_\epsilon(\lambda) \sim \exp[+\tau L^d \mu(\lambda)]$, where $\mu(\lambda) = \max_{\mathbf{J}} [G(\mathbf{J}) + \lambda \cdot \mathbf{J}]$ is the Legendre transform of the current LDF. The cumulants of the current distribution can be obtained from the derivatives of $\mu(\lambda)$ evaluated at $\lambda = 0$ (i.e., $\mu_{(n_1, \dots, n_d)}^{(n)} \equiv [\partial^{n_1} \mu(\lambda) / \partial \lambda_1^{n_1} \dots \partial \lambda_d^{n_d}]_{\lambda=0} = (\tau L^d)^{n-1} \langle \Delta J_1^{n_1} \dots \Delta J_d^{n_d} \rangle_\epsilon$ for $n = \sum n_i \geq 1$) where $\Delta J_a \equiv J_a - (1 - \delta_{n,1}) \langle J_a \rangle_\epsilon$ and $\delta_{n,m}$ is the Kronecker symbol. The IFR can be stated for the Legendre transform of the current LDF as $\mu(\lambda) = \mu[\mathcal{R}(\lambda + \epsilon) - \epsilon]$, where \mathcal{R} is any d -dimensional rotation. Using this relation in the definition of the n -th order cumulant in the limit of infinitesimal rotations, $\mathcal{R} = \mathbb{1} + \Delta\theta \mathcal{L}$, it is easy to show that

$$n_\alpha \mathcal{L}_{\beta\alpha} \mu_{(n_1 \dots n_\alpha - 1 \dots n_\beta + 1 \dots n_d)}^{(n)} + \epsilon_\nu \mathcal{L}_{\nu\mu} \mu_{(n_1 \dots n_\nu + 1 \dots n_d)}^{(n+1)} = 0, \quad [13]$$

where \mathcal{L} is any generator of d -dimensional rotations, and summation over repeated Greek indices ($\in [1, d]$) is assumed. The above hierarchy relates in a simple way cumulants of orders n and $n + 1 \forall n \geq 1$, and is valid arbitrarily far from equilibrium. As an example, Eqs. 9 and 10 above show the first two sets of relations ($n = 1, 2$) of the above hierarchy in two dimensions. In a similar way, we can explore the consequences of the IFR on the linear and nonlinear response coefficients. For that, we now expand the cumulants of the current in powers of ϵ

*Notice that ρ -dependent corrections to a constant mobility σ , which are typically irrelevant from a renormalization-group point of view (25), turn out to be essential for current fluctuations as they give rise to non-Gaussian tails in the current distribution.

$$\mu_{(n_1 \dots n_d)}^{(n)}(\epsilon) = \sum_{k=0}^{\infty} \frac{1}{k!} \sum_{\substack{k_1 \dots k_d=0 \\ \sum_i k_i=k}}^k \binom{(k)}{(n)} \chi_{(n_1 \dots n_d)}^{(k_1 \dots k_d)} \epsilon_1^{k_1} \dots \epsilon_d^{k_d} \quad [14]$$

Inserting expansion [14] into the cumulant hierarchy, Eq. 13, and matching order by order in k , we derive another interesting hierarchy for the response coefficients of the different cumulants. For $k=0$ this reads

$$n_\alpha \mathcal{L}_{\beta\alpha(n)}^{(0)} \chi_{(n_1 \dots n_\alpha - 1 \dots n_\beta + 1 \dots n_d)}^{(0 \dots 0)} = 0, \quad [15]$$

which is a symmetry relation for the equilibrium ($\epsilon=0$) current cumulants. For $k \geq 1$ we obtain

$$\sum_{\substack{k_1 \dots k_d=0 \\ \sum_i k_i=k \geq 1}}^k \left[\frac{n_\alpha}{k} \mathcal{L}_{\beta\alpha(n)}^{(k)} \chi_{(n_1 \dots n_\alpha - 1 \dots n_\beta + 1 \dots n_d)}^{(k_1 \dots k_d)} + \mathcal{L}_{\gamma\nu(n+1)}^{(k-1)} \chi_{(n_1 \dots n_\gamma - 1 \dots n_d)}^{(k_1 \dots k_\nu - 1 \dots k_d)} \right] = 0, \quad [16]$$

which relates k -order response coefficients of n -order cumulants with $(k-1)$ -order coefficients of $(n+1)$ -order cumulants. Relations [15] and [16] for

the response coefficients result from the IFR in the limit of infinitesimal rotations. For a finite rotation $\mathcal{R} = -\mathbb{1}$, which is equivalent to a current inversion, we have $\mu(\lambda) = \mu(-\lambda - 2\epsilon)$ and we may use this in the definition of response coefficients, $\binom{(n)}{(k)} \chi_{(n_1 \dots n_d)}^{(k_1 \dots k_d)} \equiv k! [\partial^{n+k} \mu(\lambda) / \partial \lambda_1^{n_1} \dots \partial \lambda_d^{n_d} \partial \epsilon_1^{k_1} \dots \partial \epsilon_d^{k_d}]_{\lambda=0=\epsilon}$, see Eq. 14, to obtain a complementary relation for the response coefficients

$$\binom{(k)}{(n)} \chi_{(n_1 \dots n_d)}^{(k_1 \dots k_d)} = k! \sum_{p_1=0}^{k_1} \dots \sum_{p_d=0}^{k_d} \frac{(-1)^{n+p} 2^p}{(k-p)!} \binom{(k-p)}{(n+p)} \chi_{(n_1+p_1 \dots n_d+p_d)}^{(k_1-p_1 \dots k_d-p_d)}, \quad [17]$$

where $p = \sum_i p_i$. A similar equation was derived in (28) from the standard fluctuation theorem, although the IFR adds further relations. All together, Eqs. 15–17 imply deep relations between the response coefficients at arbitrary orders that go far beyond Onsager's reciprocity relations and Green-Kubo formulae. As an example, we discuss in the main text some of these relations for a two-dimensional system.

ACKNOWLEDGMENTS. We acknowledge financial support from Spanish Ministerio de Ciencia e Innovación Project FIS2009-08451, the University of Granada, and Junta de Andalucía (Projects P07-FQM02725 and P09-FQM4682).

- Ritort F (2008) Nonequilibrium fluctuations in small systems: From physics to biology. *Adv Chem Phys* 137:31–123.
- Bertini L, De Sole A, Gabrielli D, Jona-Lasinio G, Landim C (2007) Stochastic interacting particle systems out of equilibrium. *J Stat Mech* P07014.
- Derrida B (2007) Non-equilibrium steady states: Fluctuations and large deviations of the density and of the current. *J Stat Mech* P07023.
- Bertini L, De Sole A, Gabrielli D, Jona-Lasinio G, Landim C (2001) Fluctuations in stationary nonequilibrium states of irreversible processes. *Phys Rev Lett* 87:040601.
- Bertini L, De Sole A, Gabrielli D, Jona-Lasinio G, Landim C (2005) Current fluctuations in stochastic lattice gases. *Phys Rev Lett* 94:030601.
- Bodineau T, Derrida B (2004) Current fluctuations in nonequilibrium diffusive systems: An additivity principle. *Phys Rev Lett* 92:180601.
- Hurtado PI, Garrido PL (2009) Test of the additivity principle for current fluctuations in a model of heat conduction. *Phys Rev Lett* 102:250601.
- Hurtado PI, Garrido PL (2010) Large fluctuations of the macroscopic current in diffusive systems: A numerical test of the additivity principle. *Phys Rev E* 81:041102.
- Bodineau T, Derrida B (2005) Distribution of current in nonequilibrium diffusive systems and phase transitions. *Phys Rev E* 72:066110.
- Lepri S, Livi R, Politi A (2003) Thermal conduction in classical low-dimensional lattices. *Phys Rep* 377:1–80.
- Dhar A (2008) Heat transport in low-dimensional systems. *Adv Phys* 57:457–537.
- Garrido PL, Hurtado PI, Nadrowski B (2001) Simple one-dimensional model of heat conduction which obeys Fourier's law. *Phys Rev Lett* 86:5486–5489.
- Hurtado PI (2006) Breakdown of hydrodynamics in a simple one-dimensional fluid. *Phys Rev Lett* 96:010601.
- Evans DJ, Cohen EGD, Morriss GP (1993) Probability of second law violations in shearing steady states. *Phys Rev Lett* 71:2401–2404.
- Gallavotti G, Cohen EGD (1995) Dynamical ensembles in nonequilibrium statistical mechanics. *Phys Rev Lett* 74:2694–2697.
- Lebowitz JL, Spohn H (1999) A Gallavotti-Cohen-type symmetry in the large deviation functional for stochastic dynamics. *J Stat Phys* 95:333–365.
- Kurchan J (1998) Fluctuation theorem for stochastic dynamics. *J Phys A-Math Gen* 31:3719–3729.
- Garrido PL, Lebowitz JL, Maes C, Spohn H (1990) Long range correlations for conservative dynamics. *Phys Rev A* 42:1954–1968.
- Bertini L, De Sole A, Gabrielli D, Jona-Lasinio G, Landim C (2007) On the long range correlations of thermodynamic systems out of equilibrium. arXiv:0705.2996.
- Gross DJ (1996) The role of symmetry in fundamental physics. *Proc Natl Acad Sci USA* 93:14256–14259.
- Luchinsky DG, McClintock PVE (1997) Irreversibility of classical fluctuations studied in analogue electrical circuits. *Nature* 389:463–466.
- Kipnis C, Marchioro C, Presutti E (1982) Heat flow in an exactly solvable model. *J Stat Phys* 27:65–74.
- Garrido PL, Gallavotti G (2007) Boundary dissipation in a driven hard disk system. *J Stat Phys* 126:1201–1207.
- Spohn H (1991) *Large Scale Dynamics of Interacting Particles* (Springer, Berlin).
- Binney JJ, Dowrick NJ, Fisher AJ, Newman MEJ (1998) *The Theory of Critical Phenomena: An Introduction to the Renormalization Group* (Oxford University Press, Oxford).
- Ellis RS (1985) *Entropy, Large Deviations, and Statistical Mechanics* (Springer, New York).
- Touchette H (2009) The large deviation approach to statistical mechanics. *Phys Rep* 478:1–69.
- Andrieux D, Gaspard P (2007) A fluctuation theorem for currents and non-linear response coefficients. *J Stat Mech* P02006.
- Pérez-Espigares C, del Pozo JJ, Garrido PL, Hurtado PI (2011) Current large deviations in a two-dimensional diffusive system. *AIP Conf Proc* 1332:204–213.
- Giardinà C, Kurchan J, Peliti L (2006) Direct evaluation of large deviation functions. *Phys Rev Lett* 96:120603.
- Hurtado PI, Garrido PL (2009) Current fluctuations and statistics during a large deviation event in an exactly-solvable transport model. *J Stat Mech* P02032.
- Collin D, et al. (2005) Verification of Crook's fluctuation theorem and recovery of RNA folding free energies. *Nature* 437:231–234.