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Unified Thermodynamic Uncertainty Relations in Linear Response

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Thermodynamic uncertainty relations (TURs) are recently established relations between the relative uncertainty of time-integrated currents and entropy production in nonequilibrium systems. For small perturbations away from equilibrium, linear response (LR) theory provides the natural framework to study generic nonequilibrium processes. Here, we use LR to derive TURs in a straightforward and unified way. Our approach allows us to generalize TURs to systems without local time-reversal symmetry, including, e.g., ballistic transport and periodically driven classical and quantum systems. We find that, for broken time reversal, the bounds on the relative uncertainty are controlled both by dissipation and by a parameter encoding the asymmetry of the Onsager matrix. We illustrate our results with an example from mesoscopic physics. We also extend our approach beyond linear response: for Markovian dynamics, it reveals a connection between the TUR and current fluctuation theorems.

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Introduction.—Central to modern statistical mechanics are general principles governing the behavior of fluctuations in systems away from thermal equilibrium. The simplest of these principles is the connection between the change of expectation values of observables in response to small perturbations and correlations of spontaneous fluctuations in equilibrium, the fluctuation-dissipation theorem (FDT) [1]. For systems arbitrarily far from equilibrium, fluctuation theorems [2–5] provide the most general characterization to date of the statistical properties of fluctuations. These general principles are not only of fundamental and conceptual importance, but also of practical benefit as they connect the hard-to-compute fluctuations in a specific system with the easier accessible constraints determined by general properties such as symmetry. For example, FDT is exploited to obtain transport coefficients from equilibrium time-correlators via Green-Kubo relations [6,7], and equilibrium free-energy differences can be recovered from nonequilibrium trajectories via the Jarzynski relation [3].

An important recent addition to the above has been the discovery of general lower bounds on the fluctuations of time-integrated currents in nonequilibrium steady states [8–14] of stochastic systems. In particular, for Markovian dynamics with local detailed balance, given a time-integrated current $J_\alpha(t)$, whose long-time average converges to $\langle J_\alpha(t)\rangle/t \to J_\alpha \neq 0$, and variance, $[\langle J_\alpha(t)^2\rangle - \langle J_\alpha(t)\rangle^2]/t$, to $D_\alpha \neq 0$, the thermodynamic uncertainty relation (TUR) [8] provides a general constraint: the squared relative uncertainty, $\varepsilon^2(t) = [\langle J_\alpha(t)^2\rangle - \langle J_\alpha(t)\rangle^2]/\langle J_\alpha(t)\rangle^2$, asymptotically obeys the inequality [8,9]

$$\varepsilon^2(t)\sigma t \to \sigma D_\alpha / J_\alpha^2 \geq 2, \tag{1}$$

where σ is the rate of entropy production. This bound implies that more precise output (smaller ε), requires more dissipation σt. The TUR (1) pertains to small deviations around the average [8,11] but was shown [9] to follow, for time homogeneous Markov processes, from a general bound also valid in the large deviation regime. Both TURs and bounds on large deviation functions have been refined and extended [10,12–15], adapted to counting observables [16], to first-passage times [16,17], generalized to finite times [18–20], to discrete time and periodic dynamics [21–23], and applied to a variety of nonequilibrium problems [24–30].

In this Letter, we consider TURs from the general point of view of linear response (LR) as applicable to systems where a nonequilibrium state (steady or periodic) arises due to small perturbations. In this regime, linear irreversible thermodynamics applies [31]: a small stationary current J_α, e.g., a particle or heat current, can be expanded in terms of affinities F_α, such as chemical potential or temperature differences, as $J_\alpha = \sum_\beta L_{\alpha\beta} F_\beta$, where the response coefficients $L_{\alpha\beta}$ form the Onsager matrix $\mathbb{L}$. Within this framework, the FDT implies $\partial J_\alpha / \partial F_\alpha = D_\alpha/2$, with $D_\alpha = 2L_{\alpha\alpha}$ describing Gaussian fluctuations near equilibrium, while the average rate of entropy production is $\sigma = \sum_\alpha F_\alpha J_\alpha$ (also valid beyond LR). The strength of LR is that it can be applied irrespective of whether the perturbed system obeys local time reversibility, with the relevant features of the dynamics encoded in the Onsager matrix. Thus, it can be used to describe ballistic transport in a magnetic field, periodically driven systems [32], and open quantum systems close to equilibrium [33].

Here, we show that, within LR, TURs can be derived in a straightforward and unified manner that accounts for systems with generic dynamical properties. In particular, we find that, for any current, i.e., for any contraction of basis currents $J_{\mathbf{c}} = \sum_\alpha c_\alpha J_\alpha$, the general TUR

$$\sigma D_{\mathbf{c}}/J_{\mathbf{c}}^2 \geq 2/(1+s_{\mathbb{L}}^2) \tag{2}$$

holds. Here, $s_{\mathbb{L}}$ is the asymmetry index of the Onsager matrix [34,35], which quantifies the extent to which the breaking of time-reversal symmetry affects response properties. We will illustrate this general TUR (2) by discussing chiral transport in a mesoscopic multiterminal conductor [36–40].

Extending our approach beyond LR, we introduce a variational principle that allows us to find the current with the smallest uncertainty. In the time-reversible case, this makes it possible to establish a connection between the TUR (1) and fluctuation theorems [2,41–44]. We also discuss generalized TURs for chiral transport beyond LR.

Linear response bounds.—Consider measuring a current $J_{\mathbf{c}}$ given by a linear combination of basis currents, $J_{\mathbf{c}} = \sum_\alpha c_\alpha J_\alpha = \mathbf{c}^T\mathbb{L}\mathbf{F}$, where $\mathbf{c}$ is a vector of real coefficients, $(\mathbf{c})_\alpha = c_\alpha$, and $\mathbf{F}$ is a vector of affinities, $(\mathbf{F})_\alpha = F_\alpha$. In LR, the fluctuations of this current around the stationary value $J_{\mathbf{c}}$ are given by $D_{\mathbf{c}} = 2\sum_{\alpha\beta} c_\alpha L_{\alpha\beta} c_\beta = 2\mathbf{c}^T\mathbb{L}\mathbf{c}$, as $\mathbb{L}$ also describes the correlations between Gaussian fluctuations of the basis currents [31]. Its relative precision (inverse of the relative uncertainty) is bounded from above by that of the current with the lowest relative uncertainty,

$$\frac{J_{\mathbf{c}}^2}{\sigma D_{\mathbf{c}}} \leq \max_{\mathbf{c}} \frac{J_{\mathbf{c}}^2}{\sigma D_{\mathbf{c}}} = \max_{\mathbf{c}} \frac{(\mathbf{c}^T\mathbb{L}\mathbf{F})^2}{2\mathbf{F}^T\mathbb{L}\mathbf{F}\mathbf{c}^T\mathbb{L}\mathbf{c}}, \tag{3}$$

where we have included the rate of entropy production $\sigma = \sum_\alpha F_\alpha J_\alpha = \mathbf{F}^T\mathbb{L}\mathbf{F}$ in the denominator [45].

For time-reversal symmetric systems, the Onsager matrix is symmetric [31]. In general, however, we have $\mathbb{L} = \mathbb{L}_S + \mathbb{L}_A$, where $\mathbb{L}_S = \mathbb{L}_S^T$ is the symmetric and $\mathbb{L}_A = -\mathbb{L}_A^T$ the antisymmetric part of $\mathbb{L}$. For any real coefficients $\mathbf{c}$, we have that current fluctuations are determined only by the symmetric part of $\mathbb{L}$, $\mathbf{c}^T\mathbb{L}\mathbf{c} = \mathbf{c}^T\mathbb{L}_S\mathbf{c}$ which, thus, must be positive semidefinite. This condition is also implied by the second law [46], as $\sigma = \mathbf{F}^T\mathbb{L}\mathbf{F} \geq 0$.

(i) Time-reversible case: First, we consider systems with a symmetric Onsager matrix, $\mathbb{L} = \mathbb{L}_S$, such as time-homogeneous Markov processes with local detailed balance. The numerator in (3) can then be written as the square of the scalar product of $\mathbb{L}^{1/2}\mathbf{c}$ and $\mathbb{L}^{1/2}\mathbf{F}$. Using the Cauchy-Schwarz inequality, $(\mathbf{c}^T\mathbb{L}\mathbf{F})^2 \leq (\mathbf{c}^T\mathbb{L}\mathbf{c})(\mathbf{F}^T\mathbb{L}\mathbf{F})$, we obtain the time-symmetric TUR

$$J_{\mathbf{c}}^2/(\sigma D_{\mathbf{c}}) \leq 1/2. \tag{4}$$

Note that (4) is saturated if $\mathbb{L}^{1/2}\mathbf{c}\|\mathbb{L}^{1/2}\mathbf{F}$. This condition requires $\mathbf{c}\|\mathbf{F}$ on the orthogonal complement of the kernel of $\mathbb{L}$, where $\mathbb{L}^{1/2}$ can be inverted. In particular, for positive $\mathbb{L}$, the only current saturating the inequality is proportional to the affinity vector $\mathbf{F}$, i.e., the entropy production [11]. For this choice of current in local detail balance dynamics, the quadratic bound on the rate function by the entropy production is also the tightest [9,13,19,47]. Notably, for $\mathbf{c}$ chosen as the νth eigenvector of the Onsager matrix, $\mathbb{L}\mathbf{c} = \lambda_\nu\mathbf{c}$, we obtain the even stronger equality

$$J_{\mathbf{c}}^2/D_{\mathbf{c}} = \lambda_\nu F_\nu^2/2, \tag{5}$$

which involves only the entropy production rate along the νth direction as $\sigma = \sum_\nu \lambda_\nu F_\nu^2$ in the diagonal basis of $\mathbb{L}$, see, also, [9].

(ii) Time-nonreversible case:Assuming that $\mathbb{L}_S$ is positive and, thus, invertible, we consider the numerator in (3) as the square of the scalar product of $\mathbb{L}_S^{1/2}\mathbf{c}$ and $\mathbb{L}_S^{-1/2}\mathbb{L}\mathbf{F}$. Via the Cauchy-Schwarz inequality, we obtain

$$\frac{J_{\mathbf{c}}^2}{\sigma D_{\mathbf{c}}} \leq \frac{\mathbf{F}^T\mathbb{L}^T\mathbb{L}_S^{-1}\mathbb{L}\mathbf{F}}{2\mathbf{F}^T\mathbb{L}_S\mathbf{F}} = \frac{1}{2} + \frac{\mathbf{F}^T\mathbb{L}_A^T\mathbb{L}_S^{-1}\mathbb{L}_A\mathbf{F}}{2\mathbf{F}^T\mathbb{L}_S\mathbf{F}}. \tag{6}$$

This inequality is saturated for

$$\mathbf{c}_{\mathrm{opt}} \propto \mathbb{L}_S^{-1}\mathbb{L}\mathbf{F} = \mathbf{F} + \mathbb{L}_S^{-1}\mathbb{L}_A\mathbf{F}, \tag{7}$$

which is generally not parallel to the affinity vector $\mathbf{F}$, as a consequence of the average currents being determined by the full $\mathbb{L}$, while the current fluctuations depend only on $\mathbb{L}_S$. Since the choice $\mathbf{c}\|\mathbf{F}$ as in (4), i.e., the entropy rate current, gives $J_{\mathbf{F}}^2/(\sigma D_{\mathbf{F}}) = 1/2$, cf. (4), the last term in (6) is necessarily positive and the inequality is weaker than in the symmetric case. This manifests the existence of reversible currents $J_\alpha^{\mathrm{rev}} = (\mathbb{L}_A\mathbf{F})_\alpha$, which, in contrast to the irreversible currents, $J_\alpha^{\mathrm{irrev}} = (\mathbb{L}_S\mathbf{F})_\alpha$, do not contribute to the total rate of entropy production or the variance of a current [35,48], thus, giving rise to more precise currents $J_{\mathbf{c}}$ that exceed the time-reversible bound (4). Furthermore, (7) and, thus, the value of the rhs of (6), can be determined from long-time averages, $\langle J_\alpha(t)\rangle/t \to (\mathbb{L}\mathbf{F})_\alpha$, and equal-time correlations, $[\langle J_\alpha(t)J_\beta(t)\rangle - \langle J_\alpha(t)\rangle\langle J_\beta(t)\rangle]/t \to 2(\mathbb{L}_S)_{\alpha\beta}$ without the need to vary the affinities, as required to recover $\mathbb{L}$ [31].

The bound (6), depends on affinities, which, in principle, can be tuned in an experimental setup. The fundamental bound on current uncertainty, which is independent from affinities, is given by

$$\frac{J_{\mathbf{c}}^2}{\sigma D_{\mathbf{c}}} \leq \frac{1}{2} + \max_{\mathbf{F}} \frac{\mathbf{F}^T \mathbb{L}_A^T \mathbb{L}_S^{-1} \mathbb{L}_A \mathbf{F}}{2\mathbf{F}^T \mathbb{L}_S \mathbf{F}}$$
$$= \frac{1}{2} + \max_{\tilde{\mathbf{F}}} \frac{\tilde{\mathbf{F}}^T \mathbb{L}_S^{-1/2} \mathbb{L}_A^T \mathbb{L}_S^{-1} \mathbb{L}_A \mathbb{L}_S^{-1/2} \tilde{\mathbf{F}}}{2\tilde{\mathbf{F}}^T \tilde{\mathbf{F}}} = \frac{1 + s_{\mathbb{L}}^2}{2}, \quad (8)$$

where $\tilde{\mathbf{F}} = \mathbb{L}_S^{1/2}\mathbf{F}$, and

$$s_{\mathbb{L}} = \|\mathbb{L}_S^{-1/2}(i\mathbb{L}_A)\mathbb{L}_S^{-1/2}\| \quad (9)$$

is the maximal eigenvalue of the (asymmetric) Hermitian matrix $\mathbb{L}_S^{-1/2}(i\mathbb{L}_A)\mathbb{L}_S^{-1/2} = \mathbb{X}$ [where $\mathbb{X}^\dagger \mathbb{X} = \mathbb{L}_S^{-1/2}\mathbb{L}_A^T \mathbb{L}_S^{-1} \mathbb{L}_A \mathbb{L}_S^{-1/2}$ appears in the second line of (8)]. Therefore, in order to saturate (8), the affinities must be chosen as $\mathbf{F}_{\text{opt}} = \mathbb{L}_S^{-1/2}\tilde{\mathbf{F}}_{\text{opt}}$ with $\tilde{\mathbf{F}}_{\text{opt}}$ belonging to the double-degenerate $s_{\mathbb{L}}^2$ eigenspace of $\mathbb{X}^\dagger\mathbb{X}$ [49].

The parameter $s_{\mathbb{L}}$ is known as the asymmetry index of the Onsager matrix $\mathbb{L}$, i.e., the minimal value of s such that $s\mathbb{L}_S + i\mathbb{L}_A$ is non-negative over complex vectors [34,35]. Since $s_{\mathbb{L}}$ depends on the Onsager matrix $\mathbb{L}$, the bound (8) [or (2)] is no longer strictly universal, in contrast to the time-reversible one (4). It is important to note that our result (8), however, still implies a semiuniversal TUR for classes of systems that admit an upper bound on the asymmetry index. Below, we demonstrate it for mesoscopic ballistic conductors, while in [50], we derive a semiuniversal TUR [51] for periodically driven mesoscopic machines [32,52–54].

Interestingly, for thermal machines with broken time-reversal symmetry, it is known that the diverging asymmetry index is necessary to achieve Carnot efficiency η_C while maintaining finite power P [35,48,56]. On the other hand, the TUR (1) has been recently related to the trade-off between power, efficiency, and constancy [25,57], implying that η_C for a time-reversible engine may be achieved at $P > 0$ provided that fluctuations of power diverge, otherwise, the power necessary vanishes, $P = 0$. Our result (2) also allows for nonvanishing power when the asymmetry index diverges, see [50], consistently with [35,48,56].

Note that the breaking of the time-symmetric TUR (4) by (6) and (8) is not a consequence of considering a particular linear combination of the basis currents. Indeed, if we fix the coefficients $\mathbf{c}$, we can maximise the precision with respect to a choice of affinities [rather than a choice of coefficients as in (6)]. This optimal affinity is

$$\mathbf{F}_{\text{opt}} \propto \mathbb{L}_S^{-1}\mathbb{L}^T\mathbf{c} = \mathbf{c} - \mathbb{L}_S^{-1}\mathbb{L}_A\mathbf{c}, \quad (10)$$

leading to a weaker relation than (4)

$$\frac{J_{\mathbf{c}}^2}{\sigma D_{\mathbf{c}}} \leq \frac{\mathbf{c}^T \mathbb{L}\mathbb{L}_S^{-1}\mathbb{L}^T\mathbf{c}}{2\mathbf{c}^T\mathbb{L}_S\mathbf{c}} = \frac{1}{2} + \frac{\mathbf{c}^T\mathbb{L}_A\mathbb{L}_S^{-1}\mathbb{L}_A^T\mathbf{c}}{2\mathbf{c}^T\mathbb{L}_S\mathbf{c}}. \quad (11)$$

Example.—As an application of our theory, we now discuss the ballistic transport of matter in mesoscopic multiterminal conductors. Such devices consist of a central junction connected to N thermochemical reservoirs with common temperature T and chemical potentials μ_α with $\alpha = 1, \ldots, N$, see Fig. 1. For nonuniform affinities $F_\alpha \equiv (\mu_\alpha - \mu)/T$, where μ is a reference chemical potential, the system is driven into a nonequilibrium steady state with finite particle currents J_α flowing in the individual terminals towards the junction. The Onsager coefficients encoding the LR properties of the conductor can be obtained from the Landauer-Büttiker formula, $L_{\alpha\beta} = \int_0^\infty dE(\delta_{\alpha\beta} - \mathcal{T}_{E\mathbf{B}}^{\alpha\beta})f_E$, which describes transport as the coherent quantum scattering of noninteracting particles [36–40]. The energy-dependent transmission coefficients $0 \leq \mathcal{T}_{E\mathbf{B}}^{\alpha\beta} \leq 1$ thereby contain the scattering amplitudes connecting incoming and outgoing single-particle waves and $f_E \equiv (2\cosh[(E-\mu)/(2T)])^{-2}$ denotes the derivative of the Fermi function. Here, the Planck and Boltzmann constants were set to 1.

For charged particles, the time-reversal symmetry of single-particle scattering processes can be broken through an external magnetic field $\mathbf{B}$. The transmission coefficients and, hence, the Onsager coefficients, are then typically not symmetric. However, the asymmetry index (9) of the Onsager matrix is still subject to the constraint [35]

$$s_{\text{MJ}} \leq \cot(\pi/N), \quad (12)$$

which follows from current conservation and gauge invariance requiring the sum rules $\sum_\alpha \mathcal{T}_{E\mathbf{B}}^{\alpha\beta} = \sum_\beta \mathcal{T}_{E\mathbf{B}}^{\alpha\beta}$ [58]. Our general result (2), thus, implies the lower bound

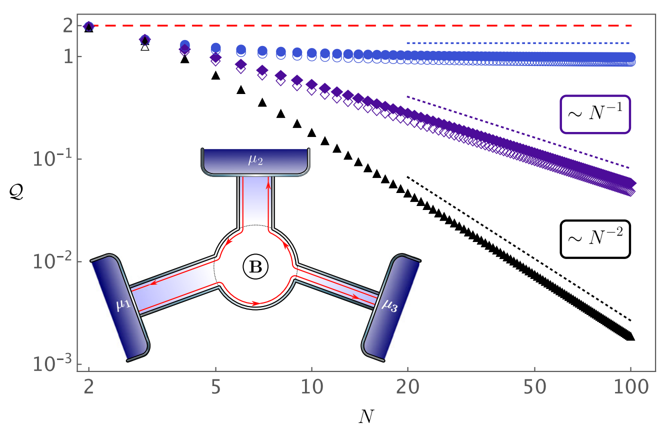

FIG. 1. Uncertainty products Q for ballistic multiterminal transport as a function of N. Inset: Setup for $N = 3$, with currents flowing along quantum Hall edge states (red lines). Main figure: Both $\mathcal{Q}_N$ for the most precise basis current (blue circles: full LR, empty beyond), and $\mathcal{Q}_{\text{lin}}$ for the optimal current (purple diamonds: full LR, empty beyond) for linear bias profile break the time-reversible TUR (1) (red dashed line). $\mathcal{Q}_{\sin}$ for sinusoidal bias (black triangles: full LR, empty beyond) saturates the LR bound (13).

$$\mathcal{Q}_{\mathbf{c}} \equiv \sigma D_{\mathbf{c}}/(J_{\mathbf{c}})^2 \geq 2\sin^2(\pi/N), \tag{13}$$

on the product of the squared relative uncertainty of any current and the rate of entropy production. We emphasize that the bound (13), independent from the potential landscape inside the junction and the strength of an external magnetic field, is valid for any mesoscopic conductor with N terminals, cf. (12) and [35].

In Fig. 1, we consider a perfectly chiral junction, which can be realized through a strong magnetic field enforcing quantum Hall edge states [59–61]. Assuming that only one edge state contributes to the transport process, the corresponding transmission coefficients are given by $\mathcal{T}^{\alpha\beta}_{E\mathbf{B}} = \delta_{\alpha(\beta-1)}$ and the Onsager coefficients read $L_{\alpha\beta} = \tau[\delta_{\alpha\beta} - \delta_{\alpha(\beta-1)}]$, where $\tau \equiv T/[1+\exp(-\mu/T)]$ [36], which corresponds to the maximal asymmetry index (12).

(a) Linear bias:First, we consider a linear bias landscape, i.e., $(\mathbf{F}_{\mathrm{lin}})_\alpha \equiv \mathcal{F}\alpha/N$, where $\mathcal{F}$ is an arbitrary constant. This choice leads to the uncertainty products $\mathcal{Q}_{\alpha<N} = N(N-1)$ and $\mathcal{Q}_N = N/(N-1)$ for the basis currents, which are bounded by 1 rather than 2, see Fig. 1 and [50]. This is due to the linear profile $\mathbf{F}_{\mathrm{lin}}$ being optimal, (10), for Nth basis current, cf. [28]. However, by combining the basis currents with the optimal coefficients for the linear profile, $(\mathbf{c}_{\mathrm{opt}})_\alpha = \mathcal{C}_N\{\alpha + [\alpha - (N+1)/2]^2 + (1-N^2)/12\}$, which follow from (7) with $\mathcal{C}_N \sim N^{-5/2}$ being the normalization factor, we obtain $J_{\mathrm{opt}} = \tau\mathcal{C}_N\mathcal{F}(N^2-1)/6$ and $D_{\mathrm{opt}} = \tau\mathcal{C}_N^2 N(N^2-1)/3$ [50]. Hence, the minimal uncertainty product $\mathcal{Q}_{\mathrm{lin}} \equiv \sigma D_{\mathrm{opt}}/(J_{\mathrm{opt}})^2 = 6/(N+1)$, vanishes for large N, see Fig. 1. Notably, due to current conservation, both $\mathcal{Q}_N$ and $\mathcal{Q}_{\mathrm{lin}}$ saturate the general bound (13) for the simplest case $N=2$, where the Onsager matrix is symmetric and (1) holds, and for the minimal nonsymmetric case $N=3$ [50].

(b) Optimal bias:To saturate the bound (13), the bias profile also has to be optimized, cf. (6) and (8). This procedure leads to the optimal affinities $(\mathbf{F}_{\mathrm{opt}})_\alpha = \mathcal{F}\cos(2\pi\alpha/N)$ with a corresponding rate of entropy production $\sigma = \mathcal{F}^2\tau N\sin^2(\pi/N)$ [50]. For this bias landscape, the uncertainty products of the basis currents increase with the number of terminals [50]. However, for the optimal current given by (7) as $(\mathbf{c}_{\mathrm{opt}})_\alpha = \mathcal{C}_N[\cos(2\pi\alpha/N) + \cot(\pi/N)\sin(2\pi\alpha/N)]$, where $\mathcal{C}_N \sim N^{-1}$ is the normalization factor, we have $J_{\mathrm{opt}} = \tau\mathcal{F}\mathcal{C}_N N$ and $D_{\mathrm{opt}} = 2\tau\mathcal{C}_N^2 N$ [50]. Thus, the minimal uncertainty product $\mathcal{Q}_{\mathrm{sin}}$ saturates the bound (13) and tends to zero as N^{-2}, see Fig. 1. We note that, for $N=3$, $\mathcal{Q}_{\mathrm{lin}} = \mathcal{Q}_{\mathrm{sin}}$, since current conservation implies the equivalence of the linear and the sinusoidal bias landscape.

Variational principle and TUR beyond linear response.—The bound (6) can be extended beyond LR using a variational principle for the relative uncertainty. To this end, first, we note that $J_{\mathbf{c}}^2/D_{\mathbf{c}} = \max_x(-x^2 D_{\mathbf{c}} + 2xJ_{\mathbf{c}})$, where the rhs attains its maximum at $x = J_{\mathbf{c}}/D_{\mathbf{c}}$. If we further maximize over $\mathbf{c}$, we get the optimal current among linear combinations of basis currents. Replacing $x\mathbf{c}$ with $\mathbf{c}$, we obtain

$$\max_{\mathbf{c}} J_{\mathbf{c}}^2/D_{\mathbf{c}} = \max_{\mathbf{c}}(-\mathbf{c}^T\mathbb{D}\mathbf{c} + 2\mathbf{c}^T\mathbf{J}). \tag{14}$$

Here, $(\mathbb{D})_{\alpha\beta} = D_{\alpha\beta}$ is the matrix of correlations between the basis currents, and $(\mathbf{J})_\alpha = J_\alpha$ the vector of average currents, which is, in general, a nonlinear function of $\mathbf{F}$. Moreover, in LR, an analogous variational principle can be obtained for the optimal choice of affinities maximizing the precision of a given current in (11) [50]. By differentiating (14) with respect to $\mathbf{c}$, we obtain the condition $\mathbb{D}\mathbf{c}_{\mathrm{opt}} = \mathbf{J}$ on the optimal coefficients $\mathbf{c}_{\mathrm{opt}}$. The relative uncertainty, $J_{\mathbf{c}}^2/D_{\mathbf{c}}$, is invariant to multiplying $\mathbf{c}$ by a scalar, so the optimality condition relaxes to

$$\mathbb{D}\mathbf{c}_{\mathrm{opt}} \propto \mathbf{J}. \tag{15}$$

If $\mathbb{D}$ is invertible, (15) leads to $\mathbf{c}_{\mathrm{opt}} \propto \mathbb{D}^{-1}\mathbf{J}$. In LR, this relation reduces to the condition (7) for saturation of (6). In general, the solution of (15) exists only if $\mathbf{J}$ is orthogonal to the kernel of $\mathbb{D}$; otherwise, the maximum of (14) is infinite and the relative uncertainty is trivially bounded from below by zero, cf. (2) [62].

In the former case, (15) implies the identity

$$\frac{1}{\mathcal{Q}_{\mathrm{opt}}} \equiv \max_{\mathbf{c}} \frac{J_{\mathbf{c}}^2}{\sigma D_{\mathbf{c}}} = \frac{\mathbf{J}^T\mathbb{D}^+\mathbf{J}}{\mathbf{F}^T\mathbf{J}}, \tag{16}$$

where $(\cdot)^+$ indicates the pseudoinverse. This relation (16) can be further formally connected to the asymmetry index in analogy to Eqs. (8) and (9), see [50,63].

(i) Time-reversible case:To the first-order beyond LR, we have $\mathbf{J} = \mathbb{L}\mathbf{F} + \delta\mathbf{J} + \mathcal{O}(\mathbf{F}^2)$ and $\mathbb{D} = 2\mathbb{L} + \delta\mathbb{D} + \mathcal{O}(\mathbf{F}^2)$, so from (16)

$$\frac{J_{\mathbf{c}}^2}{\sigma D_{\mathbf{c}}} \leq \frac{1}{2} + \frac{2\mathbf{F}^T\delta\mathbf{J} - \mathbf{F}^T\delta\mathbb{D}\mathbf{F}}{4\mathbf{F}^T\mathbb{L}\mathbf{F}} + \mathcal{O}(\mathbf{F}^2). \tag{17}$$

Both for homogeneous Markovian dynamics, and for periodically driven Markovian systems with time-reversible protocols, the first correction in (17) vanishes, as $\delta\mathbf{J} = \delta\mathbb{D}\mathbf{F}/2$ due to Gallavotti-Cohen symmetries [42,44,64]. The TUR in Eq. (4), thus, holds up to $\mathcal{O}(\mathbf{F}^2)$ for all $\mathbf{F}$ (except $\mathbf{F}$ in the kernel of $\mathbb{L}_S$). Moreover, the entropy production rate remains the optimal current, $\mathbf{c}_{\mathrm{opt}} \propto \mathbb{D}^+\mathbf{J} = \mathbf{F}/2 + \mathcal{O}(\mathbf{F}^2)$, with $\mathcal{Q}_{\mathrm{opt}} = 1/2 + \mathcal{O}(\mathbf{F}^2)$. We note that the TUR in Eq. (1) was derived beyond LR as a consequence of a quadratic bound on that rate function that also obeys the Gallavotti-Cohen symmetry [9,13,47].

(ii) Time-nonreversible case: example revisited: To explore Eq. (16) without time-reversal symmetry, we consider a chiral multiterminal junction in the nonlinear regime. For simplicity, we focus on the semiclassical limit, where the density of carriers in the conductor is low such that Pauli blocking and quantum correlations can be neglected [31]. Under this condition, the mean currents and fluctuations can be derived as $J_\alpha = \bar{\tau}(e^{F_\alpha} - e^{F_{\alpha+1}})$ and $D_{\alpha\beta} = \bar{\tau}\delta_{\alpha\beta}(e^{F_\alpha} + e^{F_{\alpha+1}}) - \bar{\tau}\delta_{\alpha(\beta-1)}e^{F_\beta} - \bar{\tau}\delta_{\beta(\alpha-1)}e^{F_\alpha}$, respectively, where $\bar{\tau} \equiv T\exp[\mu/T]$ [50]. In Fig. 1, we show how the uncertainty product $\mathcal{Q}_{\mathrm{opt}}$ for the optimal current given by (15) scales with N for linear and sinusoidal bias profiles. For the linear profile, $(\mathbf{F}_{\mathrm{lin}})_\alpha \equiv \mathcal{F}\alpha/N$, choosing the amplitude $\mathcal{F}$ to minimize $\mathcal{Q}_{\mathrm{opt}}$ leads to $\mathcal{Q}_{\mathrm{lin}} \geq \psi^* 6/(N+1)$, with an additional factor $\psi^* \simeq 0.83$ compared to LR, as occurs for the basis currents [28]; see, also, [50]. In contrast, for $N \geq 4$ and the sinusoidal bias profile $(\mathbf{F}_{\sin})_\alpha \equiv \mathcal{F}_1 \cos(2\pi\alpha/N) + \mathcal{F}_2 \sin(2\pi\alpha/N)$, the optimal amplitudes $\mathcal{F}_1$ and $\mathcal{F}_2$ are within the LR regime and the bound (13) holds; see [50] for details. As the sinusoidal bias profile is no longer guaranteed to be optimal beyond LR, only a systematic optimization of the bias profile would lead to a general TUR for ballistic conductors beyond LR, which constitutes an interesting problem for future work.

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[1] R. Kubo, M. Toda, and N. Hashitsume, *Statistical Physics II: Nonequilibrium Statistical Mechanics*, Vol. 31 (Springer, New York, 2012).
[2] G. Gallavotti and E. G. D. Cohen, Phys. Rev. Lett. **74**, 2694 (1995).
[3] C. Jarzynski, Phys. Rev. Lett. **78**, 2690 (1997).
[4] G. E. Crooks, Phys. Rev. E **60**, 2721 (1999).
[5] J. L. Lebowitz and H. Spohn, J. Stat. Phys. **95**, 333 (1999).
[6] M. S. Green, J. Chem. Phys. **22**, 398 (1954).
[7] R. Kubo, J. Phys. Soc. Jpn. **12**, 570 (1957).
[8] A. C. Barato and U. Seifert, Phys. Rev. Lett. **114**, 158101 (2015).
[9] T. R. Gingrich, J. M. Horowitz, N. Perunov, and J. L. England, Phys. Rev. Lett. **116**, 120601 (2016).
[10] M. Polettini, A. Lazarescu, and M. Esposito, Phys. Rev. E **94**, 052104 (2016).
[11] P. Pietzonka, A. C. Barato, and U. Seifert, Phys. Rev. E **93**, 052145 (2016).
[12] P. Pietzonka, A. C. Barato, and U. Seifert, J. Phys. A **49**, 34LT01 (2016).
[13] T. R. Gingrich, G. M. Rotskoff, and J. M. Horowitz, J. Phys. A **50**, 184004 (2017).
[14] U. Seifert, Physica A (Amsterdam) **504A**, 176 (2018).
[15] S. Pigolotti, I. Neri, E. Roldan, and F. Jülicher, Phys. Rev. Lett. **119**, 140604 (2017).
[16] J. P. Garrahan, Phys. Rev. E **95**, 032134 (2017).
[17] T. R. Gingrich and J. M. Horowitz, Phys. Rev. Lett. **119**, 170601 (2017).
[18] P. Pietzonka, F. Ritort, and U. Seifert, Phys. Rev. E **96**, 012101 (2017).
[19] J. M. Horowitz and T. R. Gingrich, Phys. Rev. E **96**, 020103 (2017).
[20] S. K. Manikandan and S. Krishnamurthy, J. Phys. A **51**, 11LT01 (2018).
[21] K. Proesmans and C. V. den Broeck, Europhys. Lett. **119**, 20001 (2017).
[22] D. Chiuchiù and S. Pigolotti, Phys. Rev. E **97**, 032109 (2018).
[23] A. C. Barato, R. Chetrite, A. Faggionato, and D. Gabrielli, arXiv:1806.07837.
[24] A. C. Barato and U. Seifert, J. Phys. Chem. B **119**, 6555 (2015).
[25] P. Pietzonka, A. C. Barato, and U. Seifert, J. Stat. Mech. (2016) 124004.
[26] C. Hyeon and W. Hwang, Phys. Rev. E **96**, 012156 (2017).
[27] W. Hwang and C. Hyeon, J. Phys. Chem. Lett. **9**, 513 (2018).
[28] K. Brandner, T. Hanazato, and K. Saito, Phys. Rev. Lett. **120**, 090601 (2018).
[29] A. Rosas, C. Van den Broeck, and K. Lindenberg, Phys. Rev. E **96**, 052135 (2017).
[30] L. P. Fischer, P. Pietzonka, and U. Seifert, Phys. Rev. E **97**, 022143 (2018).
[31] H. B. Callen, *Thermodynamics and an Introduction to Thermostatistics*, 2nd ed. (John Wiley & Sons, New York, 1985).
[32] K. Brandner, K. Saito, and U. Seifert, Phys. Rev. X **5**, 031019 (2015).
[33] K. Brandner and U. Seifert, Phys. Rev. E **93**, 062134 (2016).
[34] J.-P. Crouzeix and C. Gutan, Optimization **52**, 251 (2003).
[35] K. Brandner and U. Seifert, New J. Phys. **15**, 105003 (2013).
[36] M. Büttiker, Phys. Rev. B **46**, 12485 (1992).
[37] U. Sivan and Y. Imry, Phys. Rev. B **33**, 551 (1986).
[38] H. U. Baranger and A. D. Stone, Phys. Rev. B **40**, 8169 (1989).
[39] P. N. Butcher, J. Phys. Condens. Matter **2**, 4869 (1990).
[40] J. Matthews, F. Battista, D. Sánchez, P. Samuelsson, and H. Linke, Phys. Rev. B **90**, 165428 (2014).
[41] G. Gallavotti, Phys. Rev. Lett. **77**, 4334 (1996).
[42] D. Andrieux and P. Gaspard, J. Stat. Mech. (2007) P02006.
[43] A. C. Barato and R. Chetrite, J. Stat. Phys. **160**, 1154 (2015).
[44] S. Ray and A. C. Barato, Phys. Rev. E **96**, 052120 (2017).
[45] Note that the trivial lower bound on the relative precision, $0 \leq J_{\mathbf{c}}^2/\sigma D_{\mathbf{c}}$, is attained whenever $\mathbf{c}$ is orthogonal to $\mathbb{L}\mathbf{F}$ such that $J_{\mathbf{c}} = 0$.
[46] U. Seifert, Rep. Prog. Phys. **75**, 126001 (2012).
[47] C. Nardini and H. Touchette, Eur. Phys. J. B **91**, 16 (2018).
[48] K. Brandner, K. Saito, and U. Seifert, Phys. Rev. Lett. **110**, 070603 (2013).
[49] The corresponding optimal current is $\mathbf{c}_{\mathrm{opt}} = \mathbf{F}_{\mathrm{opt}} + \mathbb{L}_S^{-1}\mathbb{L}_A\mathbf{F}_{\mathrm{opt}} = \mathbb{L}_S^{-1/2}(1+\mathbb{X})\mathbf{F}_{\mathrm{opt}} = \mathbb{L}_S^{-1/2}\tilde{\mathbf{c}}_{\mathrm{opt}}$. Here, $\tilde{\mathbf{c}}_{\mathrm{opt}}$ is also a $s_{\mathbb{L}}^2$-eigenvector of $\mathbb{X}^\dagger\mathbb{X}$, since the spectrum of $\mathbb{X}$ is

reflective with respect to 0, i.e., pairs of eigenvectors with opposite eigenvalues are connected by complex conjugation.

[50] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.121.130601 for further discussions of our main results and worked out examples.

[51] Bounds for current rate functions for time-periodic Markovian systems are obtained in the very recent Ref. [23]. While the procedure there is general, the explicit bounds on current uncertainty are given only for time-independent contractions of elementary currents and, thus, exclude basis currents of extracted work or heat. In this sense, our results, here, are complementary to those in [23] and, moreover, provide the only known TUR to, e.g., heat engines without time reversal [52–55], see discussion in [50].

[52] T. Schmiedl and U. Seifert, Europhys. Lett. **81**, 20003 (2008).

[53] V. Blickle and C. Bechinger, Nat. Phys. **8**, 143 (2012).

[54] I. A. Martínez, É. Roldán, L. Dinis, D. Petrov, and R. A. Rica, Phys. Rev. Lett. **114**, 120601 (2015).

[55] H. Risken, *The Fokker-Planck Equation*, 2nd ed. (Springer, New York, 1996).

[56] G. Benenti, K. Saito, and G. Casati, Phys. Rev. Lett. **106**, 230602 (2011).

[57] P. Pietzonka and U. Seifert, Phys. Rev. Lett. **120**, 190602 (2018).

[58] Y. Blanter and M. Büttiker, Phys. Rep. **336**, 1 (2000).

[59] B. Sothmann, R. Sánchez, and A. N. Jordan, Europhys. Lett. **107**, 47003 (2014).

[60] R. Sánchez, B. Sothmann, and A. N. Jordan, Phys. Rev. Lett. **114**, 146801 (2015).

[61] R. Sánchez, B. Sothmann, and A. N. Jordan, New J. Phys. **17**, 075006 (2015).

[62] In LR, the solution of (15) exists, for arbitrary affinities $\mathbf{F}$, only if the kernel of $\mathbb{L}_S$ lies in the orthogonal complement of the range of $\mathbb{L}_A$. Note that, if the kernel of $\mathbb{L}_S$ overlaps with the range of $\mathbb{L}_A$, the asymmetry index of $\mathbb{L}$ is infinite and (8) still holds formally, see [50] for details.

[63] H. Vroylandt, D. Lacoste, and G. Verley, J. Stat. Mech. (2018) 023205.

[64] P. Gaspard, New J. Phys. **15**, 115014 (2013).