

Thermodynamic Uncertainty Relations from Exchange Fluctuation Theorems

André M. Timpanaro,^{1,*} Giacomo Guarnieri,^{2,†} John Goold,^{2,‡} and Gabriel T. Landi^{3,§}

¹Universidade Federal do ABC, 09210-580 Santo André, Brazil

²Department of Physics, Trinity College Dublin, Dublin 2, Ireland

³Instituto de Física da Universidade de São Paulo, 05314-970 São Paulo, Brazil



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Thermodynamic uncertainty relations (TURs) place strict bounds on the fluctuations of thermodynamic quantities in terms of the associated entropy production. In this Letter, we identify the tightest (and saturable) matrix-valued TUR that can be derived from the exchange fluctuation theorems describing the statistics of heat and particle flow between multiple systems of arbitrary dimensions. Our result holds for both quantum and classical systems, undergoing general finite-time nonstationary processes. Moreover, it provides bounds not only for the variances, but also for the correlations between thermodynamic quantities. To demonstrate the relevance of TURs to the design of nanoscale machines, we consider the operation of a 2-qubit SWAP engine undergoing an Otto cycle and show how our results can be used to place strict bounds on the correlations between heat and work.

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Introduction.—Over the last decades, technological developments have led to the creation of artificial meso- and nanoscopic heat engines [1,2], with applications ranging from nanojunction thermoelectrics [3] to quantum dots [4]. Understanding the fundamental principles ruling over the nonequilibrium physics of such devices is therefore one of the most sought-after challenges nowadays. One of the key features of these nonequilibrium processes is that they are always accompanied by an irreversible production of entropy, and as the systems become smaller, the fluctuations in the entropy production become significant. This requires one to treat the entropy production Σ as a random variable distributed according to a certain probability distribution $P(\Sigma)$. These distributions satisfy a set of fundamental symmetry relations, known as fluctuation theorems (FTs) [5–17], which can generally be expressed as $P(\Sigma)/\tilde{P}(-\Sigma) = e^{\Sigma}$, where $\tilde{P}(\Sigma)$ denotes the probability distribution of the time-reversed process. FTs represent a refinement of the second law of thermodynamics, which at the stochastic level is recast in the form $\langle \Sigma \rangle \geq 0$. The additional information they carry, however, can also be used to characterize systems arbitrarily far from equilibrium, which generated enormous interest across many fields of research [3,4,18–21].

More recently, another set of powerful results called thermodynamic uncertainty relations (TURs) have been discovered [22–25]. TURs impose strict restrictions on the fluctuations of thermodynamic currents (e.g., heat, particles, etc.). Letting Q denote any such *integrated* current (net charge) exchanged during an out-of-equilibrium process over some generic time interval, the TURs bound the signal-to-noise ratio (SNR) of Q according to

$$\frac{\text{Var}(Q)}{\langle Q \rangle^2} \geq \frac{2}{\langle \Sigma \rangle}, \quad (1)$$

where $\text{Var}(Q) = \langle Q^2 \rangle - \langle Q \rangle^2$ denotes the variance, $\langle Q \rangle$ is the average charge, and $\langle \Sigma \rangle$ is the average entropy production. Equation (1) expresses a trade-off between process precision, quantified by the SNR, and dissipation, quantified through the entropy production. To reduce fluctuations, one must pay the inevitable price of dissipation. This has important ramifications for the operation of microscopic autonomous engines [24], where fluctuations in the output power may be significant.

TURs were originally discovered in the context of non-equilibrium steady states of classical time-homogeneous Markov jump processes satisfying local detailed balance [22–24]. Further refinements and extensions have since been found for finite-time processes [25–27], periodically driven systems [28–30], quantum systems in linear response [31], and using geometrical arguments based on the manifold of nonequilibrium steady states [32].

A natural question that emerges is whether TURs, being inequalities, can be viewed as a consequence of FTs, just like the second law $\langle \Sigma \rangle \geq 0$. Explorations in this direction began quite recently, starting with symmetric work protocols [30,33,34] and were subsequently generalized to include measurement feedback [35,36]. In this Letter, we derive a new type of saturable TUR for FTs stemming from heat and particle exchange between multiple systems [Fig. 1(a)]. This class of problems is particularly relevant, as it encompasses microscopic autonomous engines, which can be implemented in thermoelectric devices [37,38] and are now starting to be pursued in controlled quantum platforms [39–45]. A set of charges Q_1, \dots, Q_n (energy, work, heat, particles, etc.) in this case satisfies the so-called exchange fluctuation theorems (EFTs) [11–13] (see also [14,15])

$$\frac{P(Q_1, \dots, Q_n)}{P(-Q_1, \dots, -Q_n)} = e^{\sum_i A_i Q_i}, \quad (2)$$

where A_i are thermodynamic affinities associated with each charge. The corresponding entropy production is $\Sigma = \sum_i A_i Q_i$.

As our first main result, we show that the EFT (2) implies a generalized TUR for any charge Q_i of the form

$$\frac{\text{Var}(Q_i)}{\langle Q_i \rangle^2} \geq f(\langle \Sigma \rangle), \quad (3)$$

where $f(x) = \text{csch}^2[g(x/2)]$, $\text{csch}(x)$ is the hyperbolic cosecant, and $g(x)$ is the function inverse of $x \tanh(x)$. We prove that this bound represents the tightest saturable trade-off bound for the SNR of any observable satisfying (2), given $\langle Q_i \rangle$. In fact, we are also able to provide an explicit form for the probability distribution $P(Q_1, \dots, Q_n)$ saturating (3). This is to be compared, for instance, to the bounds derived in [30,33–35], which are looser but cannot be saturated. On the other hand, a series expansion of $f(x)$ around $x = 0$ yields $f(x) \simeq 2/x - 2/3$, so that for $\langle \Sigma \rangle \ll 1$ one recovers the bound (1). The bound (1), however, does not necessarily apply to all scenarios involving the exchange fluctuation theorem and can be violated. Our bound, on the other hand, is always looser than (1) and always holds in any EFT scenario.

Our framework also allows us to go further and construct a matrix-valued TUR for the covariance matrix $C_{ij} = \text{Cov}(Q_i, Q_j) = \langle Q_i Q_j \rangle - \langle Q_i \rangle \langle Q_j \rangle$ between different charges, similar in spirit to Refs. [27,31,32]. In this case, the bound becomes

$$C - f(\langle \Sigma \rangle) \mathbf{q} \mathbf{q}^T \geq 0, \quad (4)$$

where $\mathbf{q} = (\langle Q_1 \rangle, \dots, \langle Q_n \rangle)$ and the inequality is to be interpreted as a condition on the positive semidefiniteness of the matrix on the left-hand side. This bound, therefore, not only places restrictions on the fluctuations of currents, but also on their correlations.

Equations (3) and (4) are the main results of this Letter. They hold for (i) quantum and classical systems of arbitrary dimensions and (ii) undergoing arbitrarily finite-time processes far from equilibrium. The steady-state scenario is also contemplated as a particular case in which the systems become macroscopically large [12]. Below, we start by reviewing the physical scenarios where our results are valid. We then provide the details of the proof and discuss their physical consequences. To illustrate their usefulness, we then apply them to a 2-qubit SWAP engine functioning as a nanoscale Otto cycle.

Exchange fluctuation theorem scenario.—We consider the scenario depicted in Fig. 1(a) and studied in Refs. [11–15]. An arbitrary number M of quantum systems are initially prepared in a factorized grand-canonical state

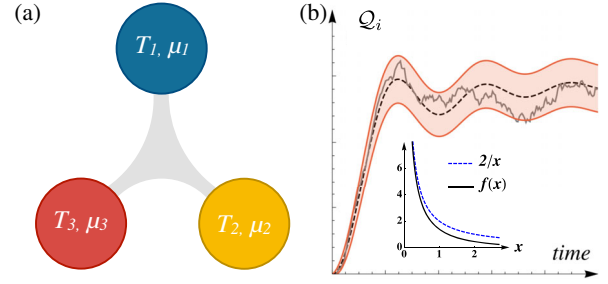


FIG. 1. (a) Exchange fluctuation theorem scenario: A system consisting of M (here $M = 3$) subsystems is allowed to interact by means of a unitary U . As a result, the subsystems will exchange energy and particles, amounting to net transferred charges (integrated currents) of energy $Q_{\mathcal{E}_i}$ and particles $Q_{\mathcal{N}_i}$. (b) For microscopic systems, any generic Q_i will be a stochastic variable and fluctuate from one repetition of the experiment to the other, represented pictorially by the jagged gray curve. This is to be contrasted with the average charge $\langle Q_i \rangle$ shown as a dashed line. The fluctuations in Q_i are represented by the variance $\text{Var}(Q_i)$, which we illustrate here by the red interval. (Inset) A plot of the function $f(x)$ on the right-hand side of Eq. (3), compared with the traditional bound $2/x$ that appears in Eq. (1).

$\rho = \prod_i Z_i^{-1} \exp[-\beta_i(\mathcal{H}_i - \mu_i \mathcal{N}_i)]$, where \mathcal{H}_i , \mathcal{N}_i are the local Hamiltonians and particle number operators, and β_i , μ_i are the inverse temperature and chemical potential of the i th subsystem [46]. The quantum systems are put in contact at time $t = 0$ up to a time τ by means of an arbitrary unitary \hat{U} incorporating the effect of all interactions between the subsystems, as well as any possible external driving. The only assumption is that the external drives are time symmetric, so that the unitary related to the time-reversed process is simply \hat{U}^\dagger . Classical systems can be treated in a similar way [11].

As a result of this time-dependent protocol, the subsystems exchange both energy and particles with each other; we denote by $Q_{\mathcal{E}_i} = \Delta \mathcal{E}_i$ and $Q_{\mathcal{N}_i} \equiv \Delta \mathcal{N}_i$ the integrated energy and particle currents during the time window $(0, \tau)$. Following [12,15], the full statistics of these quantities can be shown to satisfy the FT

$$\frac{P(Q_{\mathcal{E}_1}, \dots, Q_{\mathcal{E}_M}, Q_{\mathcal{N}_1}, \dots, Q_{\mathcal{N}_M})}{P(-Q_{\mathcal{E}_1}, \dots, -Q_{\mathcal{E}_M}, -Q_{\mathcal{N}_1}, \dots, -Q_{\mathcal{N}_M})} = e^{\sum_i \beta_i (Q_{\mathcal{E}_i} - \mu_i Q_{\mathcal{N}_i})}, \quad (5)$$

which is of the form (2).

Variations of Eq. (5) may also be naturally constructed. Consider, for instance, the particularly relevant case of $M = 2$ subsystems. Particle conservation implies that it suffices to consider the particle charge $Q_{\mathcal{N}} = \Delta \mathcal{N}_2 = -\Delta \mathcal{N}_1$ and hence work only with $P(Q_{\mathcal{E}_1}, Q_{\mathcal{E}_2}, Q_{\mathcal{N}})$. In addition, it may be of interest to change variables and use as thermodynamic quantities a heat charge $Q_H = -\Delta \mathcal{E}_1$ and a work charge $Q_W = \Delta \mathcal{E}_1 + \Delta \mathcal{E}_2$. The EFT for the joint distribution (5) then becomes [47,48]

$$\frac{P(Q_H, Q_W, Q_N)}{P(-Q_H, -Q_W, -Q_N)} = e^{\delta\beta Q_H + \beta_B Q_W + \delta\beta\mu Q_N}, \quad (6)$$

where $\delta\beta = \beta_B - \beta_A$ and $\delta\beta\mu = \beta_A\mu_A - \beta_B\mu_B$ are the corresponding affinities. This result, as stated, does not assume any form of weak coupling or strict energy conservation (i.e., in general, $\Delta\mathcal{E}_1 \neq -\Delta\mathcal{E}_2$). But if that is the case, then no work is performed and it suffices to deal with $P(Q_H, Q_N)$.

Derivation of the TUR.—We now turn to the derivation of our TUR bound. The starting point is a general joint probability distribution $P(Q_1, \dots, Q_n)$ satisfying (2). We first perform a change of variables to $\Sigma = \sum_i A_i Q_i$ and $Z = \sum_i z_i Q_i$, where z_i are a set of auxiliary variables. The corresponding probability distribution $P(\Sigma, Z) = \langle \delta(\Sigma - \sum_i A_i Q_i) \delta(Z - \sum_i z_i Q_i) \rangle$ will then have the same symmetry as Eq. (2) [49]. Namely,

$$\frac{P(\Sigma, Z)}{P(-\Sigma, -Z)} = e^\Sigma. \quad (7)$$

Our bound is now entirely based on the following simple question: for fixed $\langle \Sigma \rangle$ and $\langle Z \rangle$, what is the probability distribution $P(\Sigma, Z)$, satisfying Eq. (7), that has the smallest possible variance $\text{Var}(Z)$? We call this the minimal distribution. Our main technical contribution can then be summarized by the following theorem:

Theorem (“TUR de force”).—For fixed finite $\langle \Sigma \rangle$ and $\langle Z \rangle$, the probability distribution $P(\Sigma, Z)$ satisfying (7), with the smallest possible variance (the minimal distribution), is the distribution

$$P_{\min}(\Sigma, Z) = \frac{1}{2 \cosh(a/2)} \{ e^{a/2} \delta(\Sigma - a) \delta(Z - b) + e^{-a/2} \delta(\Sigma + a) \delta(Z + b) \}, \quad (8)$$

where the values of a and b are fixed by $\langle \Sigma \rangle = a \tanh(a/2)$ and $\langle Z \rangle = b \tanh(a/2)$.

The proof is given in the Supplemental Material [50]. We also note that a similar distribution also appears in Ref. [33]. For the minimal distribution (8), the variance of Z is given by

$$\text{Var}(Z)_{\min} = \langle Z \rangle^2 f(\langle \Sigma \rangle), \quad (9)$$

where $f(x)$ is the function discussed below Eq. (3) and $\text{Var}(Z)_{\min}$ is the variance of Z calculated with respect to P_{\min} in Eq. (8). Proving that this distribution is minimal hence implies that

$$\text{Var}(Z) \geq f(\langle \Sigma \rangle) \langle Z \rangle^2 \quad (10)$$

for any other probability distribution.

Matrix-valued TUR.—We are now in the position to complete the derivation of our TUR. The bound (10) holds

for a general combination $Z = \sum_i z_i Q_i$ of the charges, with arbitrary parameters z_i . Let us then write $\langle Z \rangle = \sum_i z_i q_i$, where $q_i = \langle Q_i \rangle$, and $\text{Var}(Z) = \sum_{ij} C_{ij} z_i z_j$, where $C_{ij} = \text{Cov}(Q_i, Q_j)$. Equation (10) can then also be written as

$$\mathbf{z}^T [C - f(\langle \Sigma \rangle) \mathbf{q} \mathbf{q}^T] \mathbf{z} \geq 0.$$

Since this must be true for any set of numbers z_i , it follows that the matrix inside the parentheses must itself be positive semidefinite. We therefore finally arrive at our main result in Eq. (4); viz., $C - f(\langle \Sigma \rangle) \mathbf{q} \mathbf{q}^T \geq 0$. The positive semidefiniteness of this matrix implies that the diagonal entries must also be non-negative. This then leads to Eq. (3).

In addition, a condition on the covariances may be obtained by using the fact that, if G is a positive semidefinite matrix, then $-\sqrt{G_{ii}G_{jj}} \leq G_{ij} \leq \sqrt{G_{ii}G_{jj}}$. Applying this to Eq. (4) immediately leads to

$$f(\langle \Sigma \rangle) q_i q_j - M_{ij} \leq C_{ij} \leq f(\langle \Sigma \rangle) q_i q_j + M_{ij}, \quad (11)$$

where $M_{ij}^2 = [\text{Var}(Q_i) - f(\langle \Sigma \rangle) q_i^2][\text{Var}(Q_j) - f(\langle \Sigma \rangle) q_j^2]$. In addition to their magnitude, particularly relevant information is also contained in the *sign* of the covariances $C_{ij} = \text{Cov}(Q_i, Q_j)$. When C_{ij} is positive (negative), values of Q_i above average imply values of Q_j above (below) average.

It is possible to find a simple criteria determining when $\text{Cov}(Q_i, Q_j)$ will have a well-defined sign (namely, the same as that of $q_i q_j$). This will occur whenever the lower and upper bounds in Eq. (11) have the same sign, which amounts to checking whether $(f q_i q_j)^2 \geq M_{ij}^2$. Using the definition of M_{ij} , one then finds

$$\frac{q_i^2}{\text{Var}(Q_i)} + \frac{q_j^2}{\text{Var}(Q_j)} \geq \frac{1}{f(\langle \Sigma \rangle)}. \quad (12)$$

If this inequality is satisfied, then it is guaranteed that $\text{sign Cov}(Q_i, Q_j) = \text{sign } q_i q_j$.

Application to a microscopic engine.—To illustrate our results, we consider the application of our bound to an engine composed of 2 qubits, with energy gaps ϵ_A and ϵ_B , interacting by means of a SWAP unitary $\hat{U} = \frac{1}{2}(1 + \hat{\sigma}_A \cdot \hat{\sigma}_B)$, where $\hat{\sigma}_i$'s are the Pauli matrices [51]. The nonresonant nature of the 2 qubits means that there will be, in general, a finite amount of work involved. As shown in Ref. [52], this work can physically be associated with the cost of turning the interaction between A and B on and off. It is not necessary to specify precisely how this takes place, however. All we need is the form of the final unitary \hat{U} .

After the qubits interact, one may reset their states by coupling them individually to two heat baths at different temperatures and allowing them to fully thermalize again [see Fig. 2(a)]. Repeating the procedure sequentially then

leads to a stroke-based engine operating at the Otto efficiency [53]. We assume A is in contact with the hot bath, so $\beta_A < \beta_B$. The change in energy of qubit A may thus be associated with the heat dumped into the hot reservoir, so we define $Q_H = -\Delta\mathcal{E}_A$. Similarly, the heat dumped to the cold reservoir is $Q_C = -\Delta\mathcal{E}_B$, whereas their mismatch is precisely the work, $W = -Q_H - Q_C = \Delta\mathcal{E}_A + \Delta\mathcal{E}_B$. The engine will thus be characterized by the stochastic variables Q_H and W . The corresponding probability distribution $P(Q_H, W)$, whose calculation details are presented in the Supplemental Material [50], will satisfy the EFT

$$\frac{P(Q_H, W)}{P(-Q_H, -W)} = e^{(\beta_B - \beta_A)Q_H + \beta_B W}, \quad (13)$$

which is clearly of the form (6), so that our basic framework applies.

Figure 2(b) shows $\langle W \rangle$, $\langle Q_H \rangle$, and $\langle \Sigma \rangle = (\beta_B - \beta_A)\langle Q_H \rangle + \beta_B \langle W \rangle$ as a function of ϵ_B/ϵ_A with fixed $\beta_A/\beta_B = 1/2$. If $(\epsilon_B/\epsilon_A) < (\beta_A/\beta_B)$, the device operates as a refrigerator, consuming work from an external agent to make heat flow from the cold to the hot bath. Instead, if $(\beta_A/\beta_B) < (\epsilon_B/\epsilon_A) < 1$, it operates as a heat engine extracting useful work ($\langle W \rangle < 0$). Finally, if $(\epsilon_B/\epsilon_A) > 1$, the device operates as an accelerator, consuming external work to increase the heat flow from hot to cold.

In Figs. 2(c) and 2(d) we present results for the fluctuations of Q_H and W , respectively. The results are compared with the bound (3) as well as the bound (1), included for comparison. As previously discussed, the bound (1) can be violated depending on the value of ϵ_B/ϵ_A . The bound (3), on the other hand, is minimal and thus can never be violated.

Finally, in Fig. 2(e) we present results for the covariance $\text{Cov}(W, Q_H)$. Studies on the correlations between thermodynamic quantities are still incipient [31]. As can be seen in the image, in both the heat engine and the refrigerator regimes, the two quantities are negatively correlated, whereas for the accelerator they become positively correlated. The covariance in this case is bounded by the interval in Eq. (11), which is represented by the two orange lines in Fig. 2(e). For all parameters of this model, in the refrigerator regime the two bounds are always negative. Equation (12), establishing the sign of $\text{Cov}(W, Q_H)$, is always satisfied only in the refrigerator regime. Thus, in this regime, $\text{Cov}(W, Q_H) < 0$ and work and heat are always anticorrelated. In the other operation regimes, such a general claim cannot be made.

Comparison with other TURs.—As discussed in the Introduction, an expansion of Eq. (3) when $\langle \Sigma \rangle \ll 1$ leads to the original TUR Eq. (1). These two bounds, however, must be compared with care, as they are derived for different physical scenarios. The original TUR (1) was obtained for time-homogeneous Markovian jump processes. Our bound,

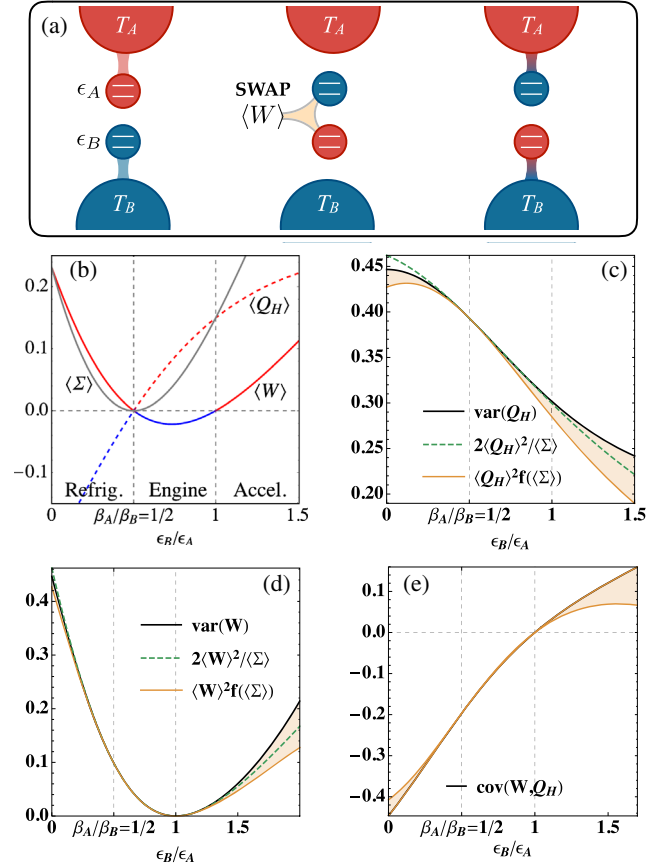


FIG. 2. Fluctuations of heat and work in a 2-qubit SWAP engine. (a) Schematic operation of the engine: 2 qubits thermalize with two baths at different temperatures. Then they are uncoupled from the baths and allowed to interact with each other by means of a SWAP operation, which produces a certain amount of work W . Repeating this procedure sequentially allows the device to operate as either a refrigerator, an engine, or an accelerator. (b) Averages of the work $\langle W \rangle$, heat to the hot bath $\langle Q_H \rangle$, and entropy production $\langle \Sigma \rangle$ as a function of ϵ_B/ϵ_A for $\beta_A/\beta_B = 1/2$. The different regimes of operation of the engine are separated by dashed vertical lines. (c) Fluctuations in the heat to the hot bath $\text{Var}(Q_H)$. The orange line represents our bound (3) and the green-dashed line represents the bound (1), included for comparison. For small values of the detuning ϵ_B/ϵ_A , (i.e., in the refrigerator regime), one can see that the bound (1) is violated (the black line lies below the green-dashed one), while (3) is always valid. (d) Same but for the fluctuations in the work $\text{Var}(W)$. (e) The correlations between heat and work, as measured by the covariance $\text{Cov}(W, Q_H)$. The two orange lines represent the bounds in Eq. (11).

on the other hand, was derived assuming only the EFT. The two scenarios do not coincide. Indeed, as shown in Fig. 2(c), the bound (1) can actually be violated in the EFT case. One situation for which the two scenarios could coincide is if the subsystems are macroscopically large. In this case, there may exist intermediate time intervals for which the exchange of energy will resemble that of a nonequilibrium steady state [54].

It is also important to compare our bound with the one derived in Refs. [30,33], which, translated into our notation, implies replacing the function $f(x)$ in Eq. (3) with

$$\frac{\text{Var}(Q_i)}{\langle Q_i \rangle^2} \geq \frac{2}{e^{\langle \Sigma \rangle} - 1}. \quad (14)$$

This bound is looser than both the original TUR (1) and our generalized TUR (3). Moreover, relevant to the present Letter, this bound was obtained by a different route than the one employed here, by means of a chain of inequalities [34]. However, as we have just proved, the bound (3) is the tightest possible bound and can only be saturated for a minimal distribution. As a consequence, the bound (14) can never be saturated. Indeed, in Ref. [35], by the same authors as in [34], the bound (14) was replaced by a bound structurally identical to Eq. (3).

Finally, we mention the connection with Ref. [32], where some of us have considered the nonequilibrium steady state of a system connected to two infinite baths, a scenario where the original TUR (1) can also be violated [55,56]. As this scenario does not satisfy an EFT, in Ref. [32] we approached the problem using the Zubarev statistical ensemble, which allowed us to show that a TUR of the form (1) also exists, but looser by a numerical factor. The two approaches therefore deal with different scenarios, but are both motivated by the same drive to generalize TURs beyond their original formulation and into the quantum regime.

Conclusions.—In this Letter, we have rigorously derived a new matrix-valued TUR solely as a consequence of EFT. This new trade-off represents the tightest bound achievable on both the signal-to-noise ratio of any integrated current and for the covariance matrix between any pair of currents. Our derivation also allowed us to explicitly find the distribution saturating this ultimate bound. This result helps to answer in the affirmative the question of whether TURs, being inequalities, can also be viewed as a consequence of fluctuation theorems, much like the second law is obtained through Jensen's inequality. It hence places an important cornerstone in the direction of understanding and controlling nonequilibrium thermodynamic processes.

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*a.timpanaro@ufabc.edu.br

†guarnieg@tcd.ie

‡gooldj@tcd.ie

§gtlandi@if.usp.br

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