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Second Law of Thermodynamics without Einstein Relation

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Materials that are constantly driven out of thermodynamic equilibrium, such as active and living systems, typically violate the Einstein relation. This may arise from active contributions to particle fluctuations which are unrelated to the dissipative resistance of the surrounding medium. We show that in these cases the widely used relation between informatic entropy production and heat dissipation does not hold. Consequently, fluctuation relations for the mechanical work, such as the Jarzynski and Crooks theorems, are invalid. We relate the breaking of the correspondence between entropy production and heat dissipation to departure from the fluctuation-dissipation theorem. We propose a temperaturelike variable that restores this correspondence and gives rise to a generalized second law of thermodynamics, whereby the dissipated heat is necessarily non-negative and vanishes at equilibrium. The Clausius inequality, Carnot maximum efficiency theorem, and relation between the extractable work and the change of free energy are recovered as well.

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Classical thermodynamics is the central theory for the possible forms of energy transfer in materials. Its many useful consequences, including the Carnot efficiency theorem and the bounds on extractable work via changes in free energy, establish nontrivial and universal relations based on a few macroscopic observables. Among these variables is the temperature, which ties microscopic statistics of equilibrium systems to their energetics, e.g., through the equipartition principle or by imposing the mean energy as a constraint for entropy maximization.

In light of its vast applicability for characterizing the behaviors of equilibrium materials, there is an ongoing effort to extend thermodynamics to systems that are constantly driven out of equilibrium, for example, self-propelled particles [1–3], sustained chemical reactions [4,5], and living systems [6]. Among the complications associated with nonequilibrium systems is the inadequacy of temperature and other variables as thermodynamic state functions [7–12].

One of the consequences of thermodynamic equilibrium, where the system's configurations are Boltzmann distributed, is the Einstein relation (ER) [13,14],

$$\mathbf{D} = k_{\mathrm{B}} T \boldsymbol{\mu}. \tag{1}$$

The diffusivity $\mathbf{D}$ and mobility $\boldsymbol{\mu}$ characterize, respectively, the strength of thermal fluctuations and dissipative attenuation of motion. In general, they may be space-dependent matrices, reflecting translational and rotational symmetry breaking. At equilibrium with a bath of temperature T, the effects of fluctuations and dissipation exactly balance [14], leading to Eq. (1). Systems away from equilibrium, such as supercooled liquids [15] and systems under external forces [8], typically do not satisfy Eq. (1) with the ambient temperature T. In such cases Eq. (1) has been used to define an effective temperature [7,10,16–18]. However, if the forces are not conservative [8,19], or when fluctuations arise from coarse graining of fast athermal or active forces [10,17,18,20–26], there is generally no relation between the two position-dependent tensors $\mathbf{D}$ and $\boldsymbol{\mu}$. Thus, in far-from-equilibrium cases such as active biopolymer networks [27–29], bacterial swarms [30,31], and driven chemical reactions [5,32], the ER is invalid, as no scalar T exists to satisfy Eq. (1). In this Letter, we propose a temperaturelike variable for systems exhibiting this severe breakdown of the Einstein relation.

One of the theories addressing nonequilibrium systems is stochastic thermodynamics [33]. By considering Langevin equations or discrete Markov chains, it establishes trajectorywise energy conservation (leading to the first law of thermodynamics) [34–36] and gives a wealth of fluctuation relations (FRs) [37], such as the Jarzynski and Crooks relations [38–40]. The latter also yields the second law of thermodynamics for the average entropy changes and heat increments. Yet, implicitly, all the aforementioned FRs and extended second law of thermodynamics have relied on the existence of a temperature, defined through the ER, to connect microscopic statistics to thermodynamic energy, mechanical and chemical work, and calorimetric heat.

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Several definitions of a nonequilibrium temperature were proposed based on generalizations of the fluctuation-dissipation theorem (FDT) and FRs in active matter [10,22,41,42], systems with memory [20,21], and anomalous diffusion [43,44]. Most of these require a thermal bath that obeys the ER. In this Letter, we adopt an alternative approach, which is not derived from extensions of FRs or FDT, to identify a temperaturelike variable directly, such that the classical-thermodynamic results are recovered.

In some cases, the ER is required to keep the extended FRs connected to physical measurables. For example, the Hatano-Sasa relation [45], an extension of the Jarzynski equality, requires Markovian dynamics only. It therefore relaxes Jarzynski's assumption of a steady-state Boltzmann distribution for a given value of the external protocol [39], thus relaxing also the ER requirement. Yet, the effective Hamiltonian and work used by this relation coincide with their actual, measurable counterparts only under those extra assumptions. Otherwise, they demand knowledge of the complete microstates' steady-state distribution, which is usually inaccessible or requires simplistic models [46]. The temperaturelike variable proposed here, arising directly from thermodynamics, does not rely on those assumptions.

We have two aims: (i) to investigate the fate of existing nonequilibrium FRs and extensions to the second law of thermodynamics once ER is violated, and (ii) to formulate a generalized, ER-independent temperature and derive the resulting alternative thermodynamic identities.

Overdamped stochastic energetics—We consider an ensemble of N interacting degrees of freedom (particles), subjected to a deterministic external time-dependent protocol Λ_t. The time-dependent stochastic microstate is an N-component vector $\mathbf{X}_t$. We couple the particles to an arbitrary medium determining the mobility $\boldsymbol{\mu}(\mathbf{x},\lambda)$ and diffusivity $\mathbf{D}(\mathbf{x},\lambda)$, such that the particles follow the overdamped Langevin equation [Eq. (2) below]; no other property of the surrounding medium is specified. The aforementioned $\mathbf{X}_t$ and Λ_t denote particular time-dependent realizations of $\mathbf{x}$ and λ. The particles are subjected to an additional external force $\mathbf{F}_{\text{ext}}(\mathbf{x},\lambda)$, which may be nonconservative and is excluded from the internal energy. We denote the total (N-component) force acting on the particles by $\mathbf{F}=\mathbf{F}_{\text{ext}}-\boldsymbol{\nabla}H$, where $H(\mathbf{x},\lambda)$ is the system's Hamiltonian. The system is driven out of equilibrium due to the nonconservative force $\mathbf{F}_{\text{ext}}$ [47] and/or violation of ER [Eq. (1)]. Thus, the system's steady-state distribution is not Boltzmann, nor can a temperature be conventionally identified.

The system follows the overdamped Langevin equation,

$$\begin{aligned}\dot{\mathbf{X}}_t = {} & \boldsymbol{\mu}(\mathbf{X}_t,\Lambda_t)\cdot\mathbf{F}(\mathbf{X}_t,\Lambda_t)+\boldsymbol{\nabla}\cdot\mathbf{D}(\mathbf{X}_t,\Lambda_t)\\ & +\mathbf{b}(\mathbf{X}_t,\Lambda_t)\cdot\boldsymbol{\eta}_t,\end{aligned}\tag{2}$$

where $\mathbf{b}$ is the noise magnitude satisfying $\mathbf{D}=\mathbf{b}\cdot\mathbf{b}^{\mathrm{T}}/2$, $\boldsymbol{\eta}_t$ is the normalized thermal noise [$\langle\boldsymbol{\eta}_t\rangle=\mathbf{0}$, $\langle\boldsymbol{\eta}_t\boldsymbol{\eta}_{t'}\rangle=\mathbf{I}\delta(t-t')$, where $\mathbf{I}$ is the identity matrix], and the equation is written in the Itô convention [48]. The "spurious drift" term $\boldsymbol{\nabla}\cdot\mathbf{D}$ ensures that Boltzmann distribution is obtained at equilibrium, i.e., in systems with a well-defined temperature, Eq. (1), under conservative forces [49,50]. The corresponding Fokker-Planck equation [51] for the microstates distribution $p(\mathbf{x},t)$ is

$$\frac{\partial p(\mathbf{x},t)}{\partial t}=-\boldsymbol{\nabla}\cdot\mathbf{J}(\mathbf{x},t),\tag{3}$$

$$\mathbf{J}=\boldsymbol{\mu}\cdot\mathbf{F}p-\mathbf{D}\cdot\boldsymbol{\nabla}p,\tag{4}$$

where $\mathbf{J}$ is the probability flux.

We follow Sekimoto's approach [34,35] to identify the internal energy, work, and heat for trajectories in generic Langevin systems. The time derivative of the Hamiltonian is [48]

$$\dot{H}_t=\boldsymbol{\nabla}H\circ\dot{\mathbf{X}}_t+\frac{\partial H}{\partial\lambda}\dot{\Lambda}_t,\tag{5}$$

where $\circ$ is the Stratonovich product [52]. $(\partial H/\partial\lambda)\mathrm{d}\Lambda_t$ is the external work necessary to change the system's internal energy over time $\mathrm{d}t$. The force $\mathbf{F}_{\text{ext}}(\mathbf{X}_t,\Lambda_t)$ acting along a displacement $\mathrm{d}\mathbf{X}_t$ during $\mathrm{d}t$ performs additional work, $\mathbf{F}_{\text{ext}}\circ\mathrm{d}\mathbf{X}_t$. Thus, the total work rate is

$$\dot{W}_t=\frac{\partial H}{\partial\lambda}\dot{\Lambda}_t+\mathbf{F}_{\text{ext}}\circ\dot{\mathbf{X}}_t.\tag{6}$$

According to the first law of thermodynamics, the heat rate is the remaining energy flow into the system,

$$\dot{Q}_t=\dot{H}_t-\dot{W}_t=-\mathbf{F}\circ\dot{\mathbf{X}}_t.\tag{7}$$

Converting the Stratonovich differential into Itô [48] and inserting Eq. (2), we find an implicit Langevin equation for the heat in terms of the solutions $\mathbf{X}_t$ of Eq. (2),

$$\dot{Q}_t=-\mathbf{F}\cdot\boldsymbol{\mu}\cdot\mathbf{F}-\boldsymbol{\nabla}\cdot(\mathbf{D}\cdot\mathbf{F})-\mathbf{F}\cdot\mathbf{b}\cdot\boldsymbol{\eta}_t.\tag{8}$$

The above is a natural adaptation of the internal energy, work, and heat to the microscale [34,35]. A similar extension of the system's entropy is less trivial. Consider the instantaneous nonequilibrium stochastic entropy [33,37], $S(\mathbf{x},t)=-\ln p(\mathbf{x},t)$, whose ensemble average is the Shannon entropy [53,57,58], $\langle S\rangle=-\int\mathrm{d}\mathbf{x}\,p(\mathbf{x},t)\ln p(\mathbf{x},t)$. The relation between classical-thermodynamic entropy and stochastic entropy is established for equilibrium states by finding the distribution p that maximizes $\langle S\rangle$ [59] (for a fixed protocol value λ). Combining the stochastic chain rule [48] and Eqs. (2)–(4) yields the time derivative of $S_t=S(\mathbf{X}_t,t)$,

$$\dot{S}_t=\boldsymbol{\nabla}\cdot\left(\frac{\mathbf{J}}{p}\right)-\frac{1}{p}\boldsymbol{\nabla}\cdot(\mathbf{D}\cdot\boldsymbol{\nabla}p)-[(\boldsymbol{\nabla}\ln p)\cdot\mathbf{b}]\cdot\boldsymbol{\eta}_t,\tag{9}$$

where the last two terms are often omitted [60] as they vanish on average.

If a well-defined temperature T exists, it is instructive to consider the thermodynamic heat dissipation, $k_B T R_t$. It quantifies how much extra calorimetric heat $\dot{Q}_t$ is wasted in an irreversible process. According to the second law of thermodynamics, the average thermodynamic dissipation $\langle \dot{R}_t \rangle$ is non-negative [61],

$$\dot{R}_t \equiv \dot{S}_t - \frac{\dot{Q}_t}{k_B T}, \qquad \langle \dot{R}_t \rangle \geq 0. \tag{10}$$

Recall that $k_B T \dot{S}_t$ is the reversible contribution to the heat. Our goal is to arrive at a similar relation in the absence of the conventional temperature T.

Informatic entropy production (IEP)—The IEP Σ_t quantifies the breaking of time-reversal symmetry in the microstates' dynamics [33]. Consider a discretized trajectory drawn from Eq. (2), $(\mathbf{X}_0, \ldots, \mathbf{X}_{K\Delta t}) = (\mathbf{x}_0, \ldots, \mathbf{x}_K)$, with arbitrarily small time step Δt. The probability distribution of obtaining this trajectory under the protocol sequence $(\lambda_0, \lambda_1, \ldots, \lambda_K)$, $\vec{\mathcal{P}} \equiv \Pr(\mathbf{x}_0, \mathbf{x}_1, \ldots, \mathbf{x}_K)$, is compared with the probability distribution of obtaining the reverted trajectory, $\overleftarrow{\mathcal{P}} \equiv \Pr(\mathbf{x}_K, \mathbf{x}_{K-1}, \ldots, \mathbf{x}_0)$, under the reverted protocol $(\lambda_K, \lambda_{K-1}, \ldots, \lambda_0)$. Explicitly [33,37],

$$\Sigma_t = \ln \frac{\vec{\mathcal{P}}}{\overleftarrow{\mathcal{P}}}. \tag{11}$$

For Markovian systems [such as Eq. (2)], it is helpful to decompose the trajectory probabilities using the infinitesimal-time propagator, $\Pr(\mathbf{x}_{k+1}, \lambda_{k+1} | \mathbf{x}_k, \lambda_k)$, as $\vec{\mathcal{P}} = p(\mathbf{x}_0, 0) \prod_{k=1}^{K} \Pr(\mathbf{x}_k, \lambda_k | \mathbf{x}_{k-1}, \lambda_{k-1})$. This decomposes the IEP rate at time $t = k\Delta t$ into

$$\begin{aligned} \dot{\Sigma}_t &= \dot{S}_t + \dot{\Omega}_t, \\ \dot{\Omega}_t &= \lim_{\Delta t \to 0} \frac{1}{\Delta t} \ln \frac{\Pr(\mathbf{x}_{k+1}, \lambda_{k+1} | \mathbf{x}_k, \lambda_k)}{\Pr(\mathbf{x}_k, \lambda_k | \mathbf{x}_{k+1}, \lambda_{k+1})}. \end{aligned} \tag{12}$$

In Eq. (12) we identify the time derivative of the entropy, $\dot{S}_t = \ln[p(\mathbf{x}_{k+1}, (k+1)\Delta t)/p(\mathbf{x}_k, k\Delta t)]/\Delta t$, given by Eq. (9). We refer to the remainder, Ω_t, as the "informatic heat" (IH). The propagator in the denominator, $\Pr(\mathbf{x}_k, \lambda_k | \mathbf{x}_{k+1}, \lambda_{k+1})$, is the exact same propagator of Eq. (2), where the jump process is reverted [i.e., the process starts at $\mathbf{x}_{k+1}$ at time $t = k\Delta t$ and ends up at $\mathbf{x}_k$ at time $(k+1)\Delta t$ under the protocol value λ_{k+1}].

Focusing on the IEP has two advantages. First, its ensemble average $\langle \Sigma_t \rangle$ is a Kullback-Leibler divergence; thus, it is non-negative [58]. Second, the dissipation coincides with the IEP, $\dot{R}_t = \dot{\Sigma}_t$, in a wide range of scenarios satisfying the Einstein relation [33]. In particular, one finds $\dot{Q}_t = -k_B T \dot{\Omega}_t$ [50] (hence the term informatic heat for Ω_t). For these cases, therefore, one does not have to resort to simplistic models for the dynamics and then employ Eq. (8) to obtain $\dot{Q}_t$. Instead, one can access $\dot{Q}_t$ from the statistics of trajectories [Eqs. (11) and (12)] without any prior knowledge of the underlying dynamics [62,63]. Clearly, the correspondences $\dot{\Omega}_t$–$\dot{Q}_t$ and $\dot{\Sigma}_t$–R_t assume the ER with its well-defined temperature.

More specifically, in Ref. [64] the IH rate is found to satisfy [65]

$$\dot{\Omega}_t = [(\boldsymbol{\mu} \cdot \mathbf{F}) \cdot \mathbf{D}^{-1}] \circ \dot{\mathbf{X}}_t. \tag{13}$$

Therefore, if $\mathbf{D}$ and $\boldsymbol{\mu}$ are not related by a scalar, the informatic $\dot{\Sigma}_t$ is no longer related to the measurable $\dot{R}_t$, even on average. Consequently, the non-negativity of $\langle \dot{R}_t \rangle$ (i.e., the second law) can no longer be established based on trajectory statistics. Below, we propose a definition of a nonequilibrium temperature that mends this discrepancy.

Nonequilibrium temperature—We return to the second law, Eq. (10). Noting that it concerns only the means of Eqs. (8) and (9), we average over the noise realizations (while using $\langle \boldsymbol{\eta}_t \rangle = \mathbf{0}$), and over the instantaneous position, where $\langle [\cdot](\mathbf{X}_t, \Lambda_t) \rangle = \int d\mathbf{x}\, p(\mathbf{x}, t) [\cdot](\mathbf{x}, \Lambda_t)$. Integration by parts gives $\langle \dot{Q}_t \rangle = -\int d\mathbf{x}\, \mathbf{J} \cdot \mathbf{F}$ and $\langle \dot{S}_t \rangle = -\int d\mathbf{x}\, \mathbf{J} \cdot \boldsymbol{\nabla} \ln p$. Let us posit that the second law of thermodynamics (10) still holds, but with a nonequilibrium temperature $\mathcal{T}$. Using the expressions for $\langle \dot{Q}_t \rangle$ and $\langle \dot{S}_t \rangle$, we express the average dissipation rate of Eq. (10) as

$$\langle \dot{R}_t \rangle = \int d\mathbf{x} \frac{\mathbf{J}}{p} \cdot \mathbf{D}^{-1} \cdot \left(\frac{\mathbf{D}}{k_B \mathcal{T}} \cdot \mathbf{F} p - \mathbf{D} \cdot \boldsymbol{\nabla} p \right). \tag{14}$$

The term in the parentheses, which depends on our choice of $\mathcal{T}$, is reminiscent of $\mathbf{J}$ [Eq. (4)].

We now impose the following physical conditions on Eq. (14): (i) To satisfy the second law, $\langle \dot{R}_t \rangle$ should be a non-negative functional of $\mathbf{J}$. (ii) To agree with Onsager's theory [66–68], the right-hand side should reduce to a quadratic form of $\mathbf{J}$ near equilibrium and turn the second-law inequality into an equality for reversible processes ($\mathbf{J} \to \mathbf{0}$). (iii) The variable $\mathcal{T}$ should be a configuration-independent scalar. The simplest choice for the dissipation functional that satisfies these three conditions is $\langle \dot{R}_t \rangle = \int d\mathbf{x} (\mathbf{J} \cdot \mathbf{D}^{-1} \cdot \mathbf{J})/p$ [69]. To complete our construction, we solve the equation $\langle \dot{R}_t \rangle = \langle \dot{S}_t \rangle - \langle \dot{Q}_t \rangle / (k_B \mathcal{T})$ for $k_B \mathcal{T}$ and find the following drive-dependent generalized temperature:

$$k_B \mathcal{T} = \frac{\int d\mathbf{x} \mathbf{J} \cdot \mathbf{F}}{\int d\mathbf{x} \mathbf{J} \cdot \mathbf{D}^{-1} \cdot (\boldsymbol{\mu} \cdot \mathbf{F})}. \tag{15}$$

We thus recovered the second law of thermodynamics for general Markovian overdamped Langevin dynamics [Eq. (2)].

Equation (15) is our central result. If ER holds, such that $\mathbf{D}^{-1} \cdot \boldsymbol{\mu} = (k_B T)^{-1}$, we readily get $\mathcal{T} = T$. Otherwise, the generalized temperature is an intricate quantity. Computing it directly from Eq. (15) is challenging. Below, we propose how to estimate it from experiments [see, e.g., Eq. (18)]. This is expected for nonequilibrium systems with arbitrarily complex $\mathbf{J}$, $\mathbf{D}$, $\boldsymbol{\mu}$, and $\mathbf{F}$. Furthermore, $\mathcal{T}$ depends on time via the protocol Λ_t affecting all these quantities. Even for constant protocol, $\mathcal{T}$ will change in time due to the instantaneous distribution $p(\mathbf{x}, t)$ entering $\mathbf{J}$. Indeed, nonequilibrium steady states generally have $\mathbf{J} \neq \mathbf{0}$, as nonconservative forces and drift create persistent fluxes across scales. Despite these caveats, Eq. (15) should play an experimentally meaningful role as a generalized temperature in nonequilibrium thermodynamics [16–18,23], as discussed below.

Consequences—First, we reestablish the connections IH–heat and IEP–dissipation, i.e., between information and thermodynamics. Substituting Eq. (2) in Eq. (13), we find

$$\begin{aligned}\dot{\Omega}_t = {} & (\boldsymbol{\mu}\cdot\mathbf{F})\cdot\mathbf{D}^{-1}\cdot(\boldsymbol{\mu}\cdot\mathbf{F}) + \boldsymbol{\nabla}\cdot(\boldsymbol{\mu}\cdot\mathbf{F}) \\ & + [(\boldsymbol{\mu}\cdot\mathbf{F})\cdot\mathbf{D}^{-1}\cdot\mathbf{b}]\cdot\boldsymbol{\eta}_t, \end{aligned} \tag{16}$$

which, combined with Eq. (9) and the relation $\dot{\Sigma}_t = \dot{S}_t + \dot{\Omega}_t$, gives the IEP rate as [70]

$$\dot{\Sigma}_t = \frac{\mathbf{J}}{p}\cdot\mathbf{D}^{-1}\cdot\frac{\mathbf{J}}{p} + \frac{2}{p}\boldsymbol{\nabla}\cdot\mathbf{J} + \left(\frac{\mathbf{J}}{p}\cdot\mathbf{D}^{-1}\cdot\mathbf{b}\right)\cdot\boldsymbol{\eta}_t. \tag{17}$$

Taking the averages of Eqs. (16) and (17), we obtain

$$k_{\mathrm{B}}\mathcal{T} = -\langle\dot{Q}_t\rangle/\langle\dot{\Omega}_t\rangle, \tag{18}$$

and

$$\langle\dot{R}_t\rangle = \langle\dot{\Sigma}_t\rangle = \int \mathrm{d}\mathbf{x}(\mathbf{J}\cdot\mathbf{D}^{-1}\cdot\mathbf{J})/p \geq 0. \tag{19}$$

Note that the correspondence holds for the means only and not for the full statistics, $\dot{Q}_t \neq -k_{\mathrm{B}}\mathcal{T}\dot{\Omega}_t$ and $\dot{R}_t \neq \dot{\Sigma}_t$. For example, the variances of the former are different, $\langle(\mathrm{d}\Omega_t)^2\rangle_{\mathrm{c}} = 2\mathrm{d}t\int(\boldsymbol{\mu}\cdot\mathbf{F})\cdot\mathbf{D}\cdot(\boldsymbol{\mu}\cdot\mathbf{F})p\mathrm{d}\mathbf{x}$, whereas $\langle(\mathrm{d}Q_t)^2\rangle_{\mathrm{c}} = 2\mathrm{d}t\int\mathbf{F}\cdot\mathbf{D}\cdot\mathbf{F}p\mathrm{d}\mathbf{x} \neq (k_{\mathrm{B}}\mathcal{T})^2\langle(\mathrm{d}\Omega_t)^2\rangle_{\mathrm{c}}$. (The subscript c denotes a cumulant.) Since the Jarzynski equality [38,39] and Crooks fluctuation theorem [40] require statistics beyond the mean, they are not restored by the generalized $\mathcal{T}$. Thus, without ER, the various FRs [37] remain related only to informatic quantities (IEP and IH) [45] and not to thermodynamics (dissipation and heat).

Another relation to examine is the one between heat of stochastic systems and the departure from FDT. Such a connection was established by Harada and Sasa [71], implicitly assuming ER and configuration-independent scalar mobility. (We recall that the ER and FDT are equivalent only in equilibrium [72].) Connections found recently for various active systems between FDT violation and the IH (e.g., Refs. [41,42]) have similarly made these two strong assumptions. As discussed above, without these assumptions the IH and thermodynamic heat are separate quantities. In addition, the assumptions underlying FDT do not apply [73]. It is then unclear which heat is connected to FDT violation—IH or thermodynamic heat? We find that neither connection can be established in the general case [73], implying that the Harada-Sasa relation breaks down as well.

Nevertheless, generalized versions of FDT and the Harada-Sasa relation can be derived in the absence of ER. Here we outline these results; see Supplemental Material for details [73]. Instead of considering the response to an impulsive linear potential gradient as was done in Ref. [71], we consider a perturbation to the force, $\delta\mathbf{F}(\mathbf{x}, s) = -\delta h_\mu(s)\boldsymbol{\mu}^{-1}(\mathbf{x}, s)\cdot\mathbf{D}(\mathbf{x}, s)\cdot\boldsymbol{\nabla}x_\mu$ at time $s < t$, in response to an external perturbation $\delta\mathbf{h}(s)$. The difference between the response of the average force to such perturbation and its correlation with the position are found to be related to the thermodynamic heat [73],

$$\langle\dot{Q}_s\rangle = -\lim_{t\to s^+}\left(\frac{\mathrm{d}}{\mathrm{d}s}\langle\mathbf{F}(\mathbf{X}_t, t)\cdot\mathbf{X}_s\rangle + \frac{\delta}{\delta\mathbf{h}(s)}\cdot\langle\mathbf{F}(\mathbf{X}_t, t)\rangle\right). \tag{20}$$

If, instead of $\mathbf{F}$, we consider the observable $\mathbf{D}^{-1}\cdot\boldsymbol{\mu}\cdot\mathbf{F}$, we obtain a similar relation for the IH,

$$\begin{aligned}\langle\dot{\Omega}_s\rangle = \lim_{t\to s^+}\Bigg(&\frac{\mathrm{d}}{\mathrm{d}s}\langle[(\boldsymbol{\mu}\cdot\mathbf{F})\cdot\mathbf{D}^{-1}](\mathbf{X}_t, t)\cdot\mathbf{X}_s\rangle \\ &+ \frac{\delta}{\delta\mathbf{h}(s)}\cdot\langle[(\boldsymbol{\mu}\cdot\mathbf{F})\cdot\mathbf{D}^{-1}](\mathbf{X}_t, t)\rangle\Bigg). \end{aligned} \tag{21}$$

Thus the IH and thermodynamic heat are related to violations of a generalized FDT for different observables. If ER holds, the two observables coincide and the classical FDT is recovered. With the additional assumption of a configuration-independent scalar mobility, the Harada-Sasa relation is recovered as well [73].

We now revisit additional classical-thermodynamic results demonstrating the relevance of the generalized temperature $\mathcal{T}$. More details regarding these consequences are found in the Supplemental Material [73]. (i) Consider a process starting and ending at steady state under the same protocol value, $\Lambda_{t_{\mathrm{f}}} = \Lambda_0$. Since $\oint_0^\infty \mathrm{d}t\langle\dot{S}_t\rangle = 0$, we obtain Clausius's inequality, $\oint_0^\infty \mathrm{d}t\langle\dot{Q}_t\rangle/(k_{\mathrm{B}}\mathcal{T}) = -\oint_0^\infty \mathrm{d}t\langle\dot{\Sigma}_t\rangle \leq 0$ [73]. (ii) Construct a Carnot-like cyclic engine consisting of an "isothermal" process (with constant $\mathcal{T}_{\mathrm{H}}$ and $\langle Q_{\mathrm{in}}\rangle > 0$), an "adiabatic" process (in which the system "cools" down to $\mathcal{T}_{\mathrm{C}}$), followed by another isothermal process (with constant $\mathcal{T}_{\mathrm{C}}$, and $\langle Q_{\mathrm{out}}\rangle < 0$), and an adiabatic process (where the system reaches $\mathcal{T}_{\mathrm{H}}$). The efficiency of such engine is $\langle -W_{\mathrm{cycle}}\rangle/\langle Q_{\mathrm{in}}\rangle = 1 - \mathcal{T}_{\mathrm{C}}/\mathcal{T}_{\mathrm{H}} + k_{\mathrm{B}}\mathcal{T}_{\mathrm{C}}\langle\Sigma_{\mathrm{cycle}}\rangle/\langle Q_{\mathrm{in}}\rangle$, where $\langle\Sigma_{\mathrm{cycle}}\rangle$ is the mean IEP during a cycle [73].

Thus, the mean IEP quantifies the cycle's irreversibility compared to a generalized Carnot efficiency, $1 - \mathcal{T}_{\mathrm{C}}/\mathcal{T}_{\mathrm{H}}$. (See also Ref. [79] in the context of nonequilibrium engines.) (iii) Define the nonequilibrium free energy, $A_t = H_t - k_{\mathrm{B}}\mathcal{T}S_t$. We show that it bounds the extractable work in isothermal (constant $\mathcal{T} > 0$) transformations, $\langle -\dot{W}_t \rangle = -(\langle \dot{A}_t \rangle + k_{\mathrm{B}}\mathcal{T}\langle \dot{\Sigma}_t \rangle) \leq -\langle \dot{A}_t \rangle$ [73].

An interesting consequence of Eq. (15) relates to a particular class of nonequilibrium systems having multiple "temperatures," e.g., a mixture of different types of particles, each of which is subjected to a different "thermostat" [80,81]. These models are relevant for mixtures of passive and active particles [82–86]. They yield a temperaturelike scalar that is a weighted average of the thermostats' temperatures. Considering a diagonal but nonscalar $\mathbf{D}^{-1} \cdot \boldsymbol{\mu}$, with different inverse temperatures as its eigenvalues, Eq. (15) is a generalization of such weighted averages. These systems may attain $\mathbf{J} = \mathbf{0}$, which results in a "0/0" ambiguity for $\mathcal{T}$. However, perturbing different global parameters amounts to taking a perturbation $\mathbf{J}$ along different axes, thus picking up different eigenvalues (inverse temperatures) of $\mathbf{D}^{-1} \cdot \boldsymbol{\mu}$. We show a simplistic example in the Supplemental Material [73].

Conclusion—We have analyzed a generic nonequilibrium Langevin system [Eq. (2)] that violates ER [Eq. (1)]. This describes ubiquitous scenarios in driven and active materials since the relation between dissipation (mobility) and fluctuations (diffusivity) is a particular consequence of the equilibrium Boltzmann distribution [14]. We have demonstrated far-reaching consequences of ER violation, which may be summarized as the detachment of informatic quantities (IH, IEP) from their physical thermodynamic counterparts (heat, dissipation) and the resulting invalidation of fluctuation theorems.

To restore the information-thermodynamics correspondence, we have imposed the second law of thermodynamics directly and identified a generalized temperature $\mathcal{T}$ [Eq. (15)], which gives all thermodynamically required properties: non-negative dissipation, zero dissipation at equilibrium, linear thermodynamic stability, and the temperature being a macroscopic (integrated) property. The generalized temperature is model-independent—with a calorimetric measurement of the heat and path sampling for the IH, $\mathcal{T}$ is found with no prior knowledge of the underlying dynamics. In particular, the variety of tools available for estimation of IEP from trajectories (e.g., sampling marginal distributions [63,87,88] or thermodynamic uncertainty relations [89,90]) will be useful, since in our construction the average IEP remains connected to the average thermodynamic dissipation. The generalized temperature does not restore the various FRs [37,38,40,71], which in the absence of ER no longer dictate the statistics of thermodynamic properties [45].

The three conditions that we used to find $\mathcal{T}$, Eq. (15), do not uniquely determine it. Within linear irreversible thermodynamics [66–68], they do guarantee a single generalized temperature, as they impose the exact leading $\mathcal{O}(J^2)$ term, $\langle \dot{R}_t \rangle = \int d\mathbf{x}(\mathbf{J} \cdot \mathbf{D}^{-1} \cdot \mathbf{J})/p$. This term is the complete dissipation functional when ER holds. If one additionally requires this dissipation functional beyond linear theory, $\mathcal{T}$ is uniquely determined by Eq. (15). The fact that $\mathcal{T}$ is the ratio between $\langle \dot{Q}_t \rangle$ (thermodynamics) and $\langle \dot{\Omega}_t \rangle$ (information) is then a useful consequence of this choice of dissipation functional.

The temperature $\mathcal{T}$ obtained here is a property of a single closed system exchanging heat with an arbitrary bath. It dictates the direction of heat flow into or out of the system based on the change in the system's entropy. The fact that the entropy production is non-negative, $\langle \dot{R}_t \rangle \geq 0$, implies a positive $\mathcal{T}$ if heat is leaving the system and a negative one if heat is entering the system at the nonequilibrium steady state. At equilibrium, there is no heat flow. It remains to be explored whether $\mathcal{T}$ also dictates the direction of heat flow between two systems in contact.

The applicability of $\mathcal{T}$ as a nonequilibrium temperature can be put to test in simple experimental scenarios. For example, one can make the mobility and diffusivity separately isotropic or anisotropic by controlling, respectively, the particle shape and athermal randomized forces. This will break ER and allow the tuning of $\mathcal{T}$ according to Eq. (15). One can then study various manifestations of the second law based on $\mathcal{T}$ and its spatial variation [18].

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