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Entropy Production in Open Systems: The Predominant Role of Intraenvironment Correlations

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We show that the entropy production in small open systems coupled to environments made of extended baths is predominantly caused by the displacement of the environment from equilibrium rather than, as often assumed, the mutual information between the system and the environment. The latter contribution is strongly bounded from above by the Araki-Lieb inequality and therefore is not time extensive, in contrast to the entropy production itself. We confirm our results with exact numerical calculations of the system-environment dynamics.

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The emergence of thermodynamic irreversibility from the reversible dynamics is one of the most important issues of thermodynamics and statistical physics. In the context of quantum (classical) systems, the problem arises from the fact that the natural candidate for the definition of the thermodynamic entropy, namely, the von Neumann (Shannon) entropy, is invariant under the unitary dynamics. In Ref. [1] this problem has been addressed by considering the joint unitary evolution of the system and the environment (which may consist of one or several baths) starting from the initially uncorrelated state $\rho_{SE}(0) = \rho_S(0) \otimes \rho_E^{\mathrm{eq}}$; the density matrices ρ_S and ρ_{SE} represent here the state of the system and the joint state of the system and the environment, respectively, whereas ρ_E^{eq} represents the Gibbs state of the environment. It was shown that the entropy production can be expressed as $\sigma \equiv D[\rho_{SE}(t)|\rho_S(t)\rho_E^{\mathrm{eq}}]$, where $D(\rho||\sigma) = \mathrm{Tr}[\rho(\ln\rho - \ln\sigma)]$ is the relative entropy (here and from here on we take $k_B = \hbar = 1$). The second law of thermodynamics $\sigma \geq 0$ results then from non-negativity of the relative entropy.

A closer look shows that the entropy production can be further decomposed into two terms [1–3],

$$\sigma = I_{SE} + D[\rho_E(t)||\rho_E^{\mathrm{eq}}], \tag{1}$$

where $I_{SE}=S_S+S_E-S_{SE}$ is the mutual information between the system and the environment and $D[\rho_E(t)||\rho_E^{\mathrm{eq}}]$ is the relative entropy between the original and the final state of the environment; here $S_i = -\mathrm{Tr}(\rho_i \ln \rho_i)$, with $i \in \{S, E, SE\}$, is the von Neumann entropy. The first term describes the system-environment correlation, whereas the second one corresponds to the displacement of the environment from equilibrium.

The natural question arising is how these terms contribute to the entropy production. It was often held [4–12] that the relative entropy $D[\rho_E(t)||\rho_E^{\mathrm{eq}}]$ is negligible for large thermal reservoirs. Based on this assumption, some recent papers even directly identified the entropy production with the mutual information between the system and the environment [6,10]. In this Letter we show, however, that in small open systems driven out of equilibrium the opposite is the case. This is because the system-environment mutual information is strongly bounded from above by the inequality [13]

$$I_{SE} \leq 2\ \min\{S_S, S_E\}, \tag{2}$$

which is a corollary of the Araki-Lieb inequality (Theorem 2 in Ref. [14]). The maximum entropy of the system is equal to $\ln N$, where N is the dimension of the Hilbert space of the system, which implies $I_{SE} \leq 2\ln N$. This bound is particularly strong in systems consisting of a few discrete energy levels, which are often studied in the context of quantum and stochastic thermodynamics [15–18]. The mutual information I_{SE}, therefore, is not a time-extensive quantity but rather saturates after a certain time, as already demonstrated numerically in Ref. [19]. In contrast, the entropy production is time extensive in systems with a continuous current flow between the baths or systems driven by some external force. We conclude, therefore, that in such a case the entropy production is related mainly to the relative entropy contribution $D[\rho_E(t)||\rho_E^{\mathrm{eq}}]$. This observation is further demonstrated by exact numerical calculations of the system-environment dynamics. We also provide a physical interpretation of the relative entropy contribution by showing that, for environments made of large baths, it may be attributed to generation of the mutual information between initially uncorrelated degrees of freedom in the environment.

General considerations.—To support our claims, let us first briefly rederive the main results of Refs. [1–3]. We consider the open quantum system described by the Hamiltonian

$$\hat{H}_{SE}(t) = \hat{H}_S(t) + \hat{H}_E + \hat{V}(t), \tag{3}$$

where $\hat{H}_S(t)$, $\hat{H}_E$, $\hat{V}(t)$ are the Hamiltonians of the system, environment, and the interaction between the system and the environment, respectively. The Hamiltonian of the environment is assumed to be time independent. For environments made of several baths, the Hamiltonian $\hat{H}_E$ can be further decomposed as $\hat{H}_E = \sum_\alpha \hat{H}_\alpha$, where $\hat{H}_\alpha$ is the Hamiltonian of the bath α.

Let us now consider the unitary evolution of the joint system starting from the initially uncorrelated state

$$\rho_{SE}(0) = \rho_S(0) \otimes \rho_E^{\mathrm{eq}} = \rho_S(0) \otimes \prod_\alpha \rho_\alpha^{\mathrm{eq}}. \tag{4}$$

Here

$$\rho_\alpha^{\mathrm{eq}} = Z_\alpha^{-1} e^{-\beta_\alpha(\hat{H}_\alpha - \mu_\alpha \hat{N}_\alpha)} \tag{5}$$

is the grand canonical Gibbs state of the bath α, where β_α and μ_α are the inverse temperature and the chemical potential of the bath, respectively. $\hat{N}_\alpha$ is the particle number operator and $Z_\alpha = \mathrm{Tr}\{\exp[-\beta_\alpha(\hat{H}_\alpha - \mu_\alpha \hat{N}_\alpha)]\}$ is the partition function. Since the unitary dynamics does not change the von Neumann entropy of the joint system, i.e., $S_{SE}(t) = S_{SE}(0)$, the mutual information between the system and the environment in the moment t can be expressed as

$$I_{SE} = \Delta S_S + \Delta S_E \geq 0, \tag{6}$$

where $\Delta S = S(t) - S(0)$. The entropy change of the environment can be further decomposed as

$$\begin{aligned}\Delta S_E &= -\mathrm{Tr}[\rho_E(t) \ln \rho_E(t)] + \mathrm{Tr}(\rho_E^{\mathrm{eq}} \ln \rho_E^{\mathrm{eq}}) \\ &= -\sum_\alpha \beta_\alpha Q_\alpha - D[\rho_E(t)||\rho_E^{\mathrm{eq}}].\end{aligned} \tag{7}$$

Here the term

$$-\sum_\alpha \beta_\alpha Q_\alpha \equiv -\mathrm{Tr}[\rho_E(t) \ln \rho_E^{\mathrm{eq}}] + \mathrm{Tr}(\rho_E^{\mathrm{eq}} \ln \rho_E^{\mathrm{eq}}) \tag{8}$$

is the heat-related contribution to the change of ΔS_E, with

$$Q_\alpha \equiv -\mathrm{Tr}\{[\rho_\alpha(t) - \rho_\alpha^{\mathrm{eq}}](\hat{H}_\alpha - \mu_\alpha \hat{N}_\alpha)\} \tag{9}$$

being the heat delivered to the system from the reservoir α within the time interval $[0, t]$. The second term

$$D[\rho_E(t)||\rho_E^{\mathrm{eq}}] \equiv \mathrm{Tr}[\rho_E(t) \ln \rho_E(t)] - \mathrm{Tr}[\rho_E(t) \ln \rho_E^{\mathrm{eq}}] \tag{10}$$

is the aforementioned relative entropy between the original and the final state of the environment.

Inserting Eq. (7) into Eq. (6) and rearranging terms, one obtains the second law of thermodynamics

$$\sigma \equiv \Delta S_S - \sum_\alpha \beta_\alpha Q_\alpha = I_{SE} + D[\rho_E(t)||\rho_E^{\mathrm{eq}}] \geq 0. \tag{11}$$

This equation relates the standard thermodynamic definition of the entropy production to the information-theoretical quantities I_{SE} and $D[\rho_E(t)||\rho_E^{\mathrm{eq}}]$. As discussed before, when the entropy production significantly exceeds $2 \ln N$ it has to be related mainly to the relative entropy contribution: $\sigma \approx \sum_\alpha -\beta_\alpha Q_\alpha \approx D[\rho_E(t)||\rho_E^{\mathrm{eq}}]$. This conclusion may be surprising because it was often held [4–12] that the term $D[\rho_E(t)||\rho_E^{\mathrm{eq}}]$ is of second order to the change of the density matrix of the environment $\Delta\rho_E = \rho_E(t) - \rho_E^{\mathrm{eq}}$ and therefore can be neglected for large thermal reservoirs. However, whereas such order-of-magnitude arguments are valid for numbers, they should be applied with care when considering complex multielement structures, such as density matrices; this is because a sum of many small contributions can still be significant. As a matter of fact, a non-negligible value of the relative entropy contribution for an extended bath has been already numerically demonstrated in Refs. [20,21], however, without noting the generality of this result; see also a similar observation of a nonvanishing contribution to ΔS_E not related to heat in Ref. [22].

Furthermore, the Araki-Lieb inequality can be easily rewritten as $\Delta S_E + S_S(t) - S_S(0) \leq 2S_S(t)$; thus $\Delta S_E \leq S_S(0) + S_S(t) \leq 2 \ln N$. This implies that the change of the von Neumann entropy of the environment is also strongly bounded from above and possibly much smaller than the heat-related contribution $-\sum_\alpha \beta_\alpha Q_\alpha$, which we later demonstrate numerically. Therefore, the change of the von Neumann entropy of the isothermal environment cannot be identified with the heat taken from the environment by the relation $\Delta S_E = -Q/T$, as done in equilibrium thermodynamics. Instead, as follows from Eq. (7), the identity $-Q/T = \Delta S_E + D[\rho_E(t)||\rho_E^{\mathrm{eq}}]$ holds, which clearly shows that the reservoir has been pushed away from equilibrium.

Relative entropy and interenvironment correlations.— This raises the question of the physical meaning of the relative entropy. Here we show that it can be, at least partially, attributed to generation of the correlation between initially uncorrelated degrees of freedom in the environment. For simplicity, let us focus on environments made of noninteracting baths described by Hamiltonians of the form

$$\hat{H}_\alpha = \sum_k \epsilon_{\alpha k} c_{\alpha k}^\dagger c_{\alpha k}, \tag{12}$$

where $c_{\alpha k}^\dagger$ ($c_{\alpha k}$) is the creation (annihilation) of the particle (boson or fermion) with the energy $\epsilon_{\alpha k}$. The thermal state of the environment can be then written as

$$\rho_E^{\mathrm{eq}} = \prod_{\alpha k} \rho_{\alpha k}^{\mathrm{eq}} = \prod_{\alpha k} Z_{\alpha k}^{-1} e^{-\beta_\alpha(\epsilon_{\alpha k}-\mu_\alpha)c_{\alpha k}^\dagger c_{\alpha k}}, \tag{13}$$

where $\rho_{\alpha k}^{\mathrm{eq}} = Z_{\alpha k}^{-1} \exp[-\beta_\alpha(\epsilon_{\alpha k}-\mu_\alpha)c_{\alpha k}^\dagger c_{\alpha k}]$ is the equilibrium density matrix of a single level with $Z_{\alpha k} = \mathrm{Tr}\{\exp[-\beta_\alpha(\epsilon_{\alpha k}-\mu_\alpha)c_{\alpha k}^\dagger c_{\alpha k}]\}$. The relative entropy of the environment can be further decomposed into two non-negative contributions as

$$D[\rho_E(t)||\rho_E^{\mathrm{eq}}] = D_{\mathrm{env}} + I_{\mathrm{env}}, \tag{14}$$

where

$$D_{\mathrm{env}} = \sum_{\alpha k} D[\rho_{\alpha k}(t)||\rho_{\alpha k}^{\mathrm{eq}}] \tag{15}$$

is the sum of relative entropies of the levels, and

$$I_{\mathrm{env}} = \sum_{\alpha k} S_{\alpha k} - S_E \tag{16}$$

is the mutual information describing the intraenvironment correlations, with $S_{\alpha k} = -\mathrm{Tr}(\rho_{\alpha k}\ln\rho_{\alpha k})$ being the von Neumann entropy of the level αk [for derivation of Eq. (14), see the Supplemental Material [23]]. We later show numerically that the second contribution may become dominant for large baths. Furthermore, in the Supplemental Material [23] we demonstrate that, at least for noninteracting systems, the relative entropy of a single level $D[\rho_{\alpha k}(t)||\rho_{\alpha k}^{\mathrm{eq}}]$ is of second order in the change of the level occupancy. Thus, the contribution D_{env} should vanish in the thermodynamic limit in which the population of each level is only weakly perturbed. However, since the order-of-magnitude analysis can be sometimes misleading (as shown before), this latter statement should be taken with care.

Example: Noninteracting resonant level.—We demonstrate our results on the example of a single fermionic level (denoted as d) coupled to two fermionic baths $\alpha \in \{L, R\}$, each containing K discrete energy levels αk with $k \in \{1, \ldots, K\}$. Generation of the mutual information in such a system has been already analyzed in Ref. [19], however, without connection to thermodynamics. The Hamiltonian of the system reads

$$\hat{H} = \epsilon_d c_d^\dagger c_d + \sum_{\alpha k} \epsilon_{\alpha k} c_{\alpha k}^\dagger c_{\alpha k} + \sum_{\alpha k} (t_{\alpha k} c_d^\dagger c_{\alpha k} + \mathrm{H.c.}), \tag{17}$$

where $i, j \in \{d, L1, \ldots, LK, R1, \ldots, RK\}$ denote the sites, $c_i^\dagger$ (c_i) is the creation (annihilation) operator of the particle on the site i, and $t_{\alpha k}$ is the tunnel coupling between the central level and the site αk. We further take the energy levels of the baths to be equally spaced, with $\epsilon_{\alpha,k+1} = \epsilon_{\alpha k} + \Delta\epsilon$, where $\Delta\epsilon = W/(K-1)$, with W being the bandwidth of the reservoirs. We also express the tunneling elements as $t_{\alpha k} = \sqrt{\Gamma_\alpha \Delta\epsilon/(2\pi)}$, where Γ_α is the coupling strength to the bath α.

The exact description of many-body systems is usually not possible since the rank of the density matrix, and thus the computational complexity, increases exponentially with the size of the system. However, since the Hamiltonian (17) is quadratic, the state of the system can be fully described by the two-point correlation matrix $\mathcal{C}$, with the matrix elements defined as [31]

$$\mathcal{C}_{ij}(t) = \mathrm{Tr}[c_i^\dagger c_j \rho_{SE}(t)]. \tag{18}$$

The rank of the correlation matrix $2K+1$ increases only linearly with the size of the system, which makes the exact description of the system numerically tractable.

The evolution of the correlation matrix is described by the equation [24]

$$\mathcal{C}(t) = e^{i\mathcal{H}t}\mathcal{C}(0)e^{-i\mathcal{H}t}, \tag{19}$$

where $\mathcal{C}(0)$ is the initial state and $\mathcal{H}$ is the matrix containing the Hamiltonian elements $\mathcal{H}_{ij}$, with $\mathcal{H}_{ii} = \epsilon_i$ and $\mathcal{H}_{\alpha k,d} = \mathcal{H}_{d,\alpha k}^* = t_{\alpha k}$ (we rederive this equation in the Supplemental Material [23]). The initial correlation matrix can be expressed as

$$\mathcal{C}(0) = \mathrm{diag}[n_d(0), n_{L1}, \ldots, n_{LK}, n_{R1}, \ldots, n_{RK}], \tag{20}$$

where $n_d(0)$ is the initial occupancy of the central level and $n_{\alpha k} = f[\beta_\alpha(\epsilon_{\alpha k}-\mu_\alpha)]$ are the thermal occupancies of the sites αk, with $f(x)$ being the Fermi distribution.

The von Neumann entropy of the subsystem $\mathcal{G}$ can be calculated as [19]

$$S_{\mathcal{G}} = -\sum_\sigma [C_\sigma \ln C_\sigma + (1-C_\sigma)\ln(1-C_\sigma)], \tag{21}$$

where C_σ are the eigenvalues of the reduced correlation matrix $\mathcal{C}_{\mathcal{G}}$ defined within the subspace $\mathcal{G}$; for example, $\mathcal{C}_E$ is the submatrix of the correlation matrix containing all the elements $\mathcal{C}_{ij}$ with $i, j \neq d$. In particular, the von Neumann entropy of a single level i equals just $S_i = -\mathcal{C}_{ii}\ln\mathcal{C}_{ii} - (1-\mathcal{C}_{ii})\ln(1-\mathcal{C}_{ii})$ and $S_S = S_d$. The heat taken from the bath α is expressed as

$$Q_\alpha = -\sum_{k=1}^{K} [\mathcal{C}_{k\alpha,k\alpha}(t) - \mathcal{C}_{k\alpha,k\alpha}(0)](\epsilon_{k\alpha}-\mu_\alpha), \tag{22}$$

which is equivalent to Eq. (9). Using Eq. (7), one may further calculate the relative entropy of the environment as $D[\rho_E(t)||\rho_E^{\mathrm{eq}}] = -\beta_L Q_L - \beta_R Q_R - \Delta S_E$.

Let us now analyze the entropy production resulting from the current flow induced by the difference of chemical potentials (voltage) $V = \mu_L - \mu_R$. In Fig. 1(a) we present the time evolution of the analyzed quantities for a given number of sites $K = 256$. One may observe that the mutual information is saturated after a time $t \approx 1$ and does not exceed $2 \ln 2$, in agreement with the Araki-Lieb inequality. As shown in Ref. [19], the bound $I_{SE} \leq 2 \ln 2$ becomes tight for high voltages V. Furthermore, the change of the von Neumann entropy of the environment ΔS_E saturates in a similar way. In contrast, the entropy production σ significantly exceeds $2 \ln 2$ and consists mostly of the relative entropy contribution $D[\rho_E(t)||\rho_E^{\mathrm{eq}}]$. For $t \approx 2$ the system reaches the asymptotic long-time state in which the entropy production, heat, and relative entropy increase monotonically. This long-time state is approximately equivalent to the steady state calculated in the thermodynamic limit; due to the finite size of the baths, the entropy production is, however, finally saturated for $t \approx 80$ (see the Supplemental Material [23]).

In Fig. 1(b) we display different contributions to the relative entropy $D[\rho_E(t)||\rho_E^{\mathrm{eq}}]$. For a given size of the bath, the dominant contribution to the relative entropy, and thus the entropy production, is the mutual information between degrees of freedom of the environment I_{env}; the term D_{env} is, however, also non-negligible and time extensive. As shown in the Supplemental Material [23], the contribution I_{env} is related both to the correlation between the baths and the intrabath correlations.

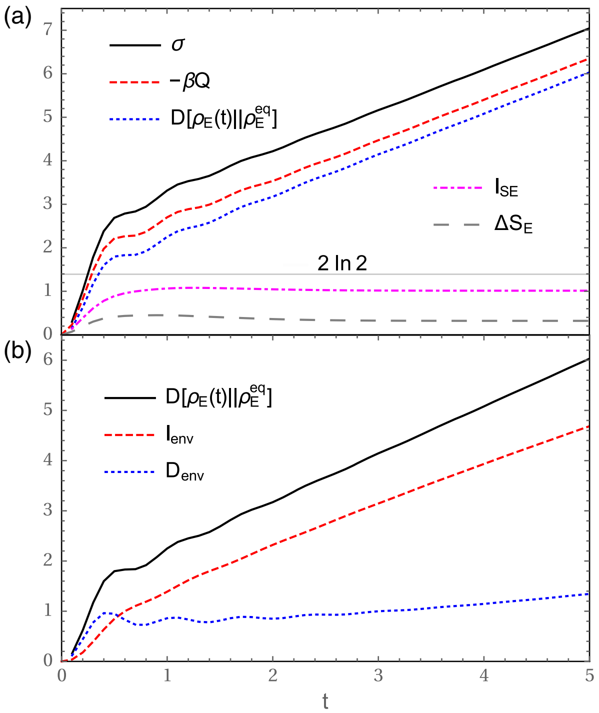

FIG. 1. (a) The entropy production σ, the heat-related contribution to the entropy production $-\beta Q = -\beta(Q_L + Q_R)$, the relative entropy of the environment $D[\rho_E(t)||\rho_E^{\mathrm{eq}}]$, the system-environment mutual information I_{SE}, and the change of the von Neumann entropy of the environment ΔS_E as a function of time for $n_d(0) = 0$, $\Gamma_L = \Gamma_R = 1/2$, $\mu_L = -\mu_R = 1$, $\beta_L = \beta_R = \beta = 3$, $\epsilon_{\alpha k} = (k-1)\Delta\epsilon - W/2$, $\Delta\epsilon = W/(K-1)$, $W = 20$, and $K = 256$. (b) Contributions to the relative entropy $D[\rho_E(t)||\rho_E^{\mathrm{eq}}]$ for parameters as in (a).

In Fig. 2, we present the dependence of the analyzed quantities on the size of the bath for a fixed time $t = 5$. One can observe a sharp transition at $K \approx 18$, which results from a crossover in the dynamics of the system: for $K \gtrapprox 18$ the entropy production grows monotonically at $t = 5$, whereas for $K \lessapprox 18$ it has already saturated due to the finite size of the bath (see the Supplemental Material [23] for details). In particular, for $K \gtrapprox 18$ the entropy production, the relative entropy of the environment, and the system-environment mutual information become independent of the number of sites. This shows that the importance of the contribution $D[\rho_E(t)||\rho_E^{\mathrm{eq}}]$ to the entropy production is not related to the size of the bath. However, the contributions of the terms D_{env} and I_{env} to the relative entropy of the environment change with the size of the bath. For $K \gtrapprox 18$ the term D_{env} decreases with the number of sites; this may be described by a power law $D_{\mathrm{env}} \propto K^{-x}$ with $x \approx 0.38$. One may expect, therefore, that in the thermodynamic limit the term D_{env} vanishes and the mutual information of the intraenvironment correlations I_{env} becomes the predominant contribution to the entropy production. This can be explained in the following way: The term D_{env} is related to the deviation of the level occupancies (diagonal elements of the correlation matrix $\mathcal{C}$) from the equilibrium ones, which becomes negligible when the baths become large (i.e., the occupancies stay thermalized during the system evolution). On the other hand, the mutual information I_{env} is associated with the creation of two-point correlations $\langle c_i^\dagger c_j \rangle$ (off-diagonal elements of the correlation matrix) that are not present in the thermal state; these correlations are responsible for the entropy production.

Final remarks.—We reemphasize that our Letter focuses on the situation when the entropy production is time

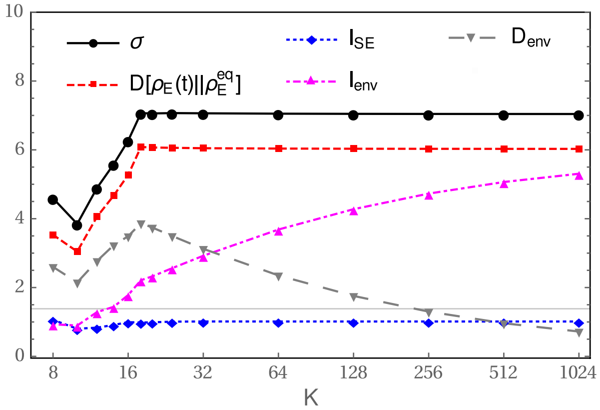

FIG. 2. The thermodynamic quantities as a function of the number of sites K for $t = 5$ and other parameters as in Fig. 1. Results denoted by points. Lines shown for eye guidance.

extensive, such that it significantly exceeds $2\ln N$ it the long-time limit. Whenever this assumption holds, the result relating the entropy production to the displacement of the environment from equilibrium $D[\rho_E(t)||\rho_E^{\mathrm{eq}}]$, rather than the system-environment mutual information I_{SE}, is general and solid (since the Araki-Lieb inequality is universally valid). Therefore, although our numerical analysis focuses on the system with a time-independent Hamiltonian, this result holds also for externally driven systems, as well as setups described within the repeated interaction framework [4,32–36] (in which the environment is made of independently prepared units interacting sequentially with the system). When, on the other hand, the entropy production is saturated at a value smaller or comparable to $2\ln N$ (which may be true, e.g., for systems undergoing thermalization [36] or short interaction quench [21]), the relative importance of the terms $D[\rho_E(t)||\rho_E^{\mathrm{eq}}]$ and I_{SE} may be not given by any general rule; instead, it may depend on details of the system-environment dynamics. This will be the topic of future study.

It is also not entirely clear whether in the thermodynamic limit the relative entropy of the environment can be always identified with the mutual information between degrees of freedom in the environment. Whereas it appears to be true for noninteracting baths composed of many levels continuously coupled to the system, the situation can be different for interacting environments or setups described within the repeated interaction framework. Therefore, our Letter may motivate further studies to better understand the mechanisms controlling the different contributions to the displacement of the environment from equilibrium. These issues, beside their fundamental importance, may also have implications for engineering environments to control dissipation.

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