Entropy production as change in observational entropy

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The thermodynamic entropy of the universe should increase in time by virtue of the second law of thermodynamics. Within the open system paradigm – where the universe is composed out of a small (quantum or classical) system coupled to one or multiple heat baths – the always positive entropy production is supposed to faithfully capture the increase in thermodynamic entropy of the universe. Indeed, recent progress has shown how to derive under very mild assumptions an always positive quantity, which resembles many features of entropy production. Unfortunately, this approach does not express the entropy production as a change in thermodynamic entropy of the universe. That is to say, the very definition of the basic thermodynamic state function measuring the entropy of the system-bath composite remained unclear.

Here, we will put forward a different approach based on the recently introduced “observational entropy” by Šafránek, Deutsch and Aguirre, which generalizes standard thermodynamic entropy to time-dependent out-of-equilibrium processes in an isolated system [1–3]. We show that the observational entropy with respect to an arbitrary measurement of the system and an energetic measurement of the bath fulfills the following key properties:

(i) Its change is always positive under even milder assumptions as used in the previous approach,
(ii) it has a clear information theoretic interpretation as the lack of knowledge about the exact microstate for a given energy $E$ of the bath, (iii) in the ideal weak-coupling case the change in observational entropy of the bath can be shown to be proportional to its change in energy divided by temperature.

Thus, the change in observational entropy provides a legitimate candidate to quantify entropy production, which is now by construction expressed as a change in a thermodynamic state function. Furthermore, we show that the change in observational entropy is typically smaller than the proposed entropy production definition of the previous approach. However, in the weak coupling limit they are quantitatively almost identical. At the end, we also verify all our general findings by using a ‘microcanonical master equation’ derived in the Markovian, weak coupling limit. This equation keeps track of energetic changes in the bath and therefore contains more information than typically used weak-coupling master equations in quantum and stochastic thermodynamics.
I. INTRODUCTION

A. Motivation

Entropy production is a central concept to quantify the irreversibility of nonequilibrium systems and dissipative structures and as such, it is of central importance to understand many situations encountered in biology, chemistry, physics and engineering [4]. Within the open system paradigm, where a system exchanges energy and entropy with its environment, entropy production $\Sigma$ is typically expressed as

$$\Sigma = \Delta S_S + \Delta S_{env} \geq 0.$$  

(1)

Here, $\Delta S_S$ ($\Delta S_{env}$) measures the change in thermodynamic entropy of the system (environment). Positivity of entropy production and the second law of nonequilibrium thermodynamics become equivalent statements then. Furthermore, if the environment is composed out of several ideal heat baths $\nu$, the change in thermodynamic entropy of the environment can be expressed as $\Delta S_{env} = \sum_\nu \Delta S_\nu$ with $\Delta S_\nu = -\beta_\nu Q_\nu$. Here, $Q_\nu$ is the heat flow from bath $\nu$ into the system and the proportionality constant is the inverse temperature $\beta_\nu$ of the bath. Then, entropy production takes on the familiar form

$$\Sigma = \Delta S_S - \sum_\nu \beta_\nu Q_\nu \geq 0.$$  

(2)

In this paper we are interested in small systems $S$, which can be dominated by fluctuations and might show quantum effects. Also within this scenario the notion of entropy production plays an essential role, e.g., to quantify the efficiency of molecular motors or quantum heat engines in stochastic and quantum thermodynamics (see, e.g., Refs. [5–7] for introductions). Conventionally, positivity of entropy production is derived from an effective master or Fokker-Planck equation, which describes the dynamics of the system after tracing out the bath under various assumptions [5–8]. While this reassures the thermodynamic consistency of the derived master or Fokker-Planck equation, it is somewhat unsatisfactory as it is not clear how entropy production emerges from the underlying microscopic (i.e., Hamiltonian) dynamics of the system and the baths.

This paper claims to provide a valid and satisfactory microscopic derivation of entropy production for a large class of open system scenarios. Indeed, as we will review in the next section, recent progress in nonequilibrium statistical mechanics has almost provided a satisfactory answer to this question. But one important point was left open: so far it was impossible to find a definition for a thermodynamic state function, which, first, describes the entropy of the system-bath composite (the ‘universe’) and, second, whose change is equal to the entropy production. In that respect previous derivations failed to show that entropy production measures the change in thermodynamic entropy of the universe.

B. Previous derivations of positivity of entropy production

Deriving the laws of thermodynamics from an underlying microscopic (i.e., Hamiltonian) picture is a central theme of statistical mechanics since its beginning. Here, we briefly review the approach by Esposito, Lindenberg and Van den Broeck [9] where a microscopic (i.e., Hamiltonian) derivation of the second law of nonequilibrium thermodynamics for a driven system in contact with multiple heat baths was provided. In case of a single heat bath, this result was independently derived in Ref. [10], see also Ref. [11] for an early derivation in the classical case where a system acts sequentially with different baths prepared at different temperatures. Similar work and extensions of this approach can be found in Refs. [12–18]. Furthermore, related work showed how to derive ‘dissipation inequalities’ on a Hamiltonian basis for a system in contact with a single heat bath [11, 19–23]. In this approach it is assumed that the system starts in equilibrium and can relax back to it after the process has finished. While employing similar mathematical manipulations, this approach is, strictly speaking, less general than the one that will be reviewed here which takes explicitly the nonequilibrium nature of the initial and final system states into account. Also multiple baths are commonly not treated there, but see Ref. [23]. For completeness, let us also mention an alternative approach for cyclic Hamiltonian dynamics (i.e., where the initial and final Hamiltonian are the same) based on the notion of complete passivity [24]. For the rest of this section we will focus on the derivation in Ref. [9]. We will also establish notation here and illustrate the problem with this approach, which we will eventually overcome later on. We remark that we choose a quantum mechanical notation throughout this paper, but the corresponding classical manipulations are analogous.

Within the standard paradigm of open system theory, we will assume that the dynamics of the universe are modeled by the Hamiltonian $H_{tot}(\lambda_t) = H_S(\lambda_t) + H_{SB} + H_B$. Here, $H_S(\lambda_t)$ is the system Hamiltonian with $\lambda_t$ some externally
specified driving protocol (e.g., a changing electric field). Furthermore, $H_B$ describes the bath Hamiltonian and $H_{SB}$ the interaction between system and bath. The present approach can be generalized to the case of multiple heat baths and a driven interaction Hamiltonian, but for ease of presentation we refrain from doing so in this section. Now, the only assumption we will add is that the initial state of the universe is given by

$$\rho_{\text{tot}}(0) = \rho_S(0) \otimes \pi_B,$$

where $\rho_S(0)$ is arbitrary and $\pi_B = e^{-\beta H_B} / Z_B$ with $Z_B = \text{tr}_B \{ e^{-\beta H_B} \}$ denotes the canonical equilibrium state of the bath. This assumption is essential for the following and conventionally used in open system theory [8]. An extension to correlated initial states is also possible for a single heat bath [25, 26], see also Refs. [27–29] for related ‘dissipation inequalities’ and Ref. [24] for cyclic Hamiltonian processes. In this paper, however, we will exclusively focus on initially uncorrelated states.

Now, let us denote by $S_{vN}(\rho) = -\text{tr} \{ \rho \ln \rho \}$ the von Neumann entropy of the state $\rho$. Then, since the global dynamics are entropy-preserving, we find immediately with the help of Eq. (3) that

$$\Delta S_{vN}[\rho_S(t)] + \Delta S_{vN}[\rho_B(t)] = I[\rho_{SB}(t)] \geq 0,$$

where $\Delta S_{vN}[\rho(t)] = S_{vN}[\rho(t)] - S_{vN}[\rho(0)]$ denotes the change in von Neumann entropy and $I[\rho_{SB}(t)] = S_{vN}[\rho_S(t)] + S_{vN}[\rho_B(t)] - S_{vN}[\rho_{SB}(t)] \geq 0$ is the always positive mutual information. It is tempting to view Eq. (4) already as the entropy production: at least it is always positive and given by a change in a state function, namely the sum of the local von Neumann entropies. Indeed, if the ‘bath’ itself is microscopically small, it makes sense to identify the heat flux as in Eq. (4) formally takes on the conventional form of entropy production, compare with Eq. (2) in case of a single heat bath. Unfortunately, for a mesoscopic or macroscopic heat bath this does not provide a satisfactory resolution as the mutual information can always be bounded by $I[\rho_{SB}] \leq 2Q \ln \dim(H_S)$, where $H_S$ denotes the Hilbert space of the system $S$, the factor $2Q$ is 2 for quantum systems and 1 for classical systems, and we assumed that (quite naturally) $\dim(H_S) \leq \dim(H_B)$. Thus, e.g., for a two-level system the entropy production would be bounded from above for all times by $2Q \ln 2$, which is clearly in general not the case. As a counterexample it suffices to consider, e.g., a driven system subjected to a laser field which dissipates energy into its environment. The entropy production in this case should rather scale extensively with time. This point was recently emphasized in Ref. [31].

Therefore, one employs a second important step by noting the exact identity $\Delta S_{vN}[\rho_B(t)] = \beta \Delta E_B - D[\rho_B(t)\|\pi_B]$, where $\Delta E_B = \text{tr}_B \{ H_B[\rho_B(t) - \rho_B(0)] \}$ is the change in bath energy and $D[\rho\|\sigma] = \text{tr} \{ \rho [\ln \rho - \ln \sigma] \} \geq 0$ is the always positive relative entropy. Then, one identifies

$$\hat{\Sigma} = \Delta S_S + \beta \Delta E_B = I[\rho_{SB}(t)] + D[\rho_B(t)\|\pi_B] \geq 0$$

as the entropy production, here denoted by $\hat{\Sigma}$ to distinguish it from our approach put forward later on. Upon further identifying (minus) the change in bath energy with the heat flux into the system, $Q = -\Delta E_B$, one obtains $\hat{\Sigma} = \Delta S_S - \beta Q$ as in Eq. (2) for a single heat bath. It should be noted, however, that the correct identification of heat is subtle outside the limit of a weakly coupled Markovian bath, compare, e.g., with the discussion in the classical case [25, 29, 32–34] or various approaches in quantum thermodynamics [26, 35–41]. Therefore, we will mainly use the notation $\Delta E_B$ here.

Let us summarize the picture so far: Eq. (5) proposes a definition of entropy production, which

(i) is positive for arbitrary system-bath dynamics and arbitrary system and bath sizes based solely on assumption (3),

(ii) has a natural information theoretic interpretation\(^1\), and

(iii) has the conventional form as in phenomenological nonequilibrium dynamics [Eq. (2)], whereas attention has to be paid to the point that the identification with heat is only valid in the weak coupling limit.\(^2\)

While these achievements are remarkable, they leave open the question what is the definition of thermodynamic entropy for the universe $S_{\text{univ}}$, whose change equals the entropy production. If $\hat{\Sigma}$ were the correct definition of entropy production, then the thermodynamic entropy of the universe at time $t$ must be

$$S_{\text{univ}}(t) = S_{vN}[\rho_S(t)] + \beta \text{tr}_B \{ H_B \rho_B(t) \} + c$$

$$= S_{vN}[\rho_S(t)] + S_{vN}[\rho_B(t)] + D[\rho_B(t)\|\pi_B] + \hat{c}$$

$$= S_{vN}[\rho_S(t)] + S_{vN}[\rho_B(t)] + D[\rho_B(t)\|\rho_B(0)] + \hat{c}$$

\(^1\) Namely as deviation of the actual bath state $\rho_B(t)$ from the equilibrium state $\pi_B$ as measured by the relative entropy. See also Ref. [31] for a more specific discussion in that direction.

\(^2\) In the weak coupling limit $\Delta E_B$ can be rigourously linked to the heat exchanges $Q$ (known, e.g., from a weak coupling master equation) by use of the two point measurement approach, see Ref. [42].
such that $\Delta S_{\text{univ}}(t) = \hat{\Sigma}$. Here $c$ and $\hat{c}$ are arbitrary additive constants, which do not depend on time. Obviously, the first line seems to be a rather awkward definition for thermodynamic entropy. However, the second line reveals that this definition is identical to the local von Neumann entropies plus the relative entropy $D[\rho_B(t)\|\pi_B]$. While looking more reasonable, this definition depends on the initial Gibbs state of the bath (and therefore on the initial temperature), which is also unacceptable: the formal definition of a thermodynamic entropy should not depend on such details. For the third line we noticed that $\pi_B = \rho_B(0)$ such that we can get rid of the formal dependence on $\pi_B$ in the definition, but unfortunately the so defined entropy would no longer be a state function.

The goal of this paper is exactly to overcome this deficiency, while retaining the important properties (i) to (iii) above. This requires us to put forward a different approach to the problem.

C. Observational entropy

Our notion of entropy for the universe, whose change we will identify with the entropy production, is based on the recently introduced notion of “observational” (or “coarse grained”) entropy by Šafránek, Deutsch and Aguirre [1–3]. They claim that observational entropy provides a satisfactory generalization of standard thermodynamic entropy to time-dependent out-of-equilibrium processes for arbitrary isolated quantum and classical systems. What was not investigated in Ref. [1–3], however, is the question whether observational entropy also applies to the open system paradigm considered here (i.e., where the isolated ‘system’ is divided into a small system of primary interest and the surrounding heat baths).

The basic idea of observational entropy is relatively easy explained in the classical case. Suppose we perform a set of measurements on a thermodynamic system described by partitioning the phase space $\Gamma$ into different cells according to some coarse-graining procedure. Thus, each coarse graining $C_\alpha = \{C_{\alpha_i}\}$ partitions the phase space into non-overlapping regions corresponding to one measurement outcome, i.e., $\Gamma = \cup_i C_{\alpha_i}$ and $C_{\alpha_i} \cap C_{\alpha_j} = \emptyset$ for $i \neq j$. Here, $\alpha \in \{1, \ldots, M\}$ denotes the different measurements (e.g., position of particles, energy, magnetization, etc.). The thermodynamic entropy of a system measured in such a way is then postulated to be

$$S_{\text{obs}} \equiv - \sum_{C_1, \ldots, C_M} p_{C_1, \ldots, C_M} \ln \frac{p_{C_1, \ldots, C_M}}{V_{C_1, \ldots, C_M}},$$

where $p_{C_1, \ldots, C_M}$ denotes the probability to obtain the joint measurement outcome $(C_1, \ldots, C_M)$ and $V_{C_1, \ldots, C_M}$ describes the number of possible microstates associated to it.

A couple of remarks are in order. First, as also noted in Ref. [1–3], not every observational entropy is thermodynamically meaningful. Its usefulness depends crucially on the chosen observables. Second, for a ‘fine-grained’ measurement of all positions and momenta of all particles, we reproduce the Gibbs-Shannon entropy. On the other hand, if we measure only the energy $E$, and if this is a conserved quantity, then we get the Boltzmann (surface but not volume!) entropy $\ln V_E$ back. Finally, a quantum extension by using projection operators in Hilbert space is possible but more complicated as the order of the measurements now plays a role [1, 2]. Šafránek et al. argue, however, that for typical situations encountered in thermodynamics, the effect of the non-commutativity of the measurements becomes very small. In our case we will actually choose commuting measurements, namely one measurement performed on the system Hilbert space and one on the bath Hilbert space. The construction of observational entropy is then straightforward, see below.

D. Outline

The rest of this paper is organized as follows: In Sec. II we will present our choice of measurements, which defines the observational entropy. Then, we will show that the change of observational entropy provides a legitimate candidate for entropy production by verifying the points (i) to (iii) above for arbitrary open system dynamics as in Sec. IB. We will also quantitatively compare our definition of entropy production with the previous approach and the case of multiple heat baths is treated in Sec. IID. Additional generally valid observations are discussed in Sec. IIE. While the results in Sec. II are very general, they are necessarily also a bit abstract. Hence, in Sec. III we will derive a ‘microcanonical master equation’ for a system weakly coupled to a Markovian bath. This master equation takes explicitly into account changes of the bath energy. Within this important approximation we will then see that we are also able to verify all the above properties. Finally, we will conclude our findings in Sec. IV.
II. GENERAL PICTURE

Our definition of thermodynamic entropy for the system-bath setup requires two measurements: an arbitrary (but fine-grained) measurement of the system described by a set of rank-1 projectors $|s⟩⟨s|$, $s \in \{1, \ldots, \dim \mathcal{H}_S\}$, and a measurement of the bath energy described by a set of projection operators $Π_{E,δ} = \sum_{E_i\in(E-δ,E)}Π_{E_i,0}$. Here, $δ$ denotes a suitable width of the measured energy window (to be further specified in Sec. II B) and $Π_{E_i,0}$ projects on a sharp energy $E_i$ such that $H_BΠ_{E_i,0} = E_iΠ_{E_i,0}$ (notice that $Π_{E_i,0}$ is not a rank-1 projector in case of exact degeneracies).

For an arbitrary system-bath state $ρ_{SB}$ the average post measurement state is given by

$$\sum_{s,E} |s⟩⟨s|Π_{E,δ}ρ_{SB}Π_{E,δ}|s⟩ = \sum_{s,E} p_{sE}|s⟩ ⊗ ρ_B(s,E). \tag{8}$$

Here, $p_{sE} = ⟨s|\text{tr}_B\{Π_{E,δ}ρ_{SB}\}|s⟩$ denotes the probability to obtain measurement outcome $(s, E)$ and the state of the bath conditioned on that outcome is $ρ_B(s, E) = ⟨s|Π_{E,δ}ρ_{SB}Π_{E,δ}|s⟩/p_{sE}$. Note that the average post measurement state (8) has lost all quantum correlations, but is in general classically correlated. Now, the observational entropy [1–3] becomes in this case

$$S_{\text{obs}} = -\sum_{s,E} p_{sE} \ln \frac{p_{sE}}{V_{E,δ}}, \tag{9}$$

where $V_{E,δ} = \text{tr}_B\{Π_{E,δ}\}$ is the number of microstates in the bath with respect to a given energy window $(E - δ, E]$.

The claim of this paper is now that $Σ ≡ \Delta S_{\text{obs}}(t) = S_{\text{obs}}(t) - S_{\text{obs}}(0)$ is the entropy production of a system (perhaps subjected to a time-dependent driving $λ_t$) coupled to a single heat bath (multiple baths are treated in Sec. II D) by verifying the points (i) to (iii) from above. As in Sec. II B we will need an additional condition on the initial system-bath state, which is, however, milder than Eq. (3). Furthermore, we remark that our approach is in some sense close to the two-point measurement approach [42] with the difference that we do not have to perfectly measure the energy of the bath, see Sec. II B.

Before verifying the points (i) to (iii) in Sec. II C, we start with two general identities in Sec. II A followed by a discussion in Sec. II B of how small we have to choose the width $δ$ in an experiment such that all theoretical claims of this paper remain true even if $δ > 0$.

A. Two general identities

We start by noting that for any system-bath state $ρ_{SB}$

$$S_{\text{obs}} = S_{\text{Sh}}(p_s) + S_{\text{obs}}^{E_{\text{obs}}} - I(p_{sE}). \tag{10}$$

Here, $S_{\text{Sh}}(p_s) ≡ -\sum_s p_s \ln p_s$ is the Shannon entropy of the probability distribution $p_s = \sum_E p_{sE}$ and

$$S_{\text{obs}}^{E_{\text{obs}}} ≡ -\sum_E p_E \ln \frac{p_E}{V_{E,δ}} \tag{11}$$

can be interpreted as the observational entropy of the bath alone with respect to an energy measurement. Finally, $I(p_{sE}) = \sum_{s,E} p_{sE} \ln(p_{sE}/p_{sE})$ is the classical mutual information between the measurement results of the system state and the bath energy.

The second identity concerns only $S_{\text{obs}}^{E_{\text{obs}}}$. For an arbitrary bath state $ρ_B$ let $ρ_B(E) ≡ Π_{E,δ}ρ_BΠ_{E,δ}/p_E$ be the post-measurement state of the bath conditioned on outcome $E$ ignoring the measurement result $s$. This state is obtained with probability $p_E = \sum_s p_{sE} = \text{tr}_B\{Π_{E,δ}ρ_B\}$. Furthermore, we introduce the microcanonical equilibrium state $ρ_{\text{mic}}(E) = Π_{E,δ}/V_{E,δ}$ with respect to a given energy $E$. Then, a straightforward calculation reveals

$$S_{\text{obs}}^{E_{\text{obs}}} = S_{\text{Sh}} \left[ \sum_E p_E ρ_B(E) \right] + \sum_E p_E D[ρ_B(E)||ρ_{\text{mic}}(E)], \tag{12}$$

where we used the identity

$$S_{\text{Sh}}(p_E) + \sum_i p_E S_{\text{Sh}}[ρ_B(E)] = S_{\text{Sh}} \left[ \sum_E p_E ρ_B(E) \right], \tag{13}$$
which holds since the states $\rho_B(E)$ are supported on orthogonal subspaces, see Theorem 11.10 in Ref. [43]. Equation (12) tells us that the observational entropy of the bath is identical to the fine-grained (von Neumann) entropy of the average post-measurement state plus the additional ignorance (measured by the relative entropy) due to not knowing the precise microstate of the bath for a given energy $E$.

B. Initial state and ideal measurement limit

For the moment, let us consider the initial state (3), generalizations are discussed in Sec. II E. The crucial ingredient to show positivity of the second law in our approach is that, initially at time $t = 0$, the observational entropy of the bath $S_{\text{obs}}^{E_B}(0)$ must coincide with the von Neumann entropy of the average post-measurement state $\sum_E p_E(0) \rho_B(E, 0)$. This means that, using Eq. (12), the following expression must vanish:

$$S_{\text{obs}}^{E_B}(0) - S_{\text{vN}} \left[ \sum_E p_E(0) \rho_B(E, 0) \right] = \sum_E p_E(0) D[\rho_B(E, 0) \parallel \rho_{\text{mic}}(E)] \quad (14)$$

For a Gibbs state of the bath we have

$$p_E(0) = \pi_E \equiv \sum_{E, i \in (E-\delta, E]} \frac{e^{-\beta E_i V_{E,0}}}{Z_B}, \quad \rho_B(E, 0) = \pi_B(E) \equiv \sum_{E, i \in (E-\delta, E]} \Pi_{E,i,0} e^{-\beta E_i} p_E(0) Z_B. \quad (15)$$

Here, the projector $\Pi_{E,i,0}$ has rank $V_{E,i,0} = \text{tr}_{B} \{\Pi_{E,i,0}\}$ (which is greater than one in case of exact degeneracies). Using this, it becomes clear that Eq. (14) can be written as

$$S_{\text{obs}}^{E_B}(0) - S_{\text{vN}}(\pi_B) = \sum_{E} \sum_{E, i \in (E-\delta, E]} e^{-\beta E_i} V_{E,0} \ln \frac{e^{-\beta E_i} V_{E,0}}{Z_B} \sum_{E_j \in (E-\delta, E]} e^{-\beta E_i} V_{E,j,0}, \quad (16)$$

which vanishes in the ideal theoretical limit $\delta \to 0$. For finite $\delta$ we proceed by looking at the argument of the logarithm,

$$\sum_{E_j \in (E-\delta, E]} e^{-\beta E_i} V_{E,j,0} = \sum_{E_j \in (E-\delta, E]} e^{-\beta E_i} V_{E,j,0}, \quad (17)$$

where we defined $E_j = E - \delta_j$. Now, if the bath is macroscopically large, we expect that we can replace the sums by integrals and by using the mean value theorem for integration, we end up with

$$\frac{e^{\beta \delta \xi} V_{E,0}}{\int_0^\delta dx e^{\beta \xi} V_{E-x,0}} = \frac{e^{\beta \delta \xi} V_{E,\delta}}{\int_0^\delta dx V_{E-x,0}} = e^{\beta (\delta_i - \xi)} = 1 + \beta (\delta_i - \xi) + O(\beta \xi^2) \quad (18)$$

with $\xi \in (0, \delta)$ and we used $\int_0^\delta dx V_{E-x,0} = V_{E,\delta}$. Thus, in the limit

$$\beta \delta \ll 1 \quad (19)$$

the observational entropy practically coincides with the von Neumann entropy for a thermal state. Experimentally, this condition has to be met in order to ensure positivity of entropy production if the bath is initially in a Gibbs state. In the following we will assume that the energy window $\delta$ is chosen small enough such that Eq. (19) holds and we will henceforth simply denote $V_E = V_{E,\delta}$ and $\Pi_E = \Pi_{E,\delta}$.

C. Verifying points (i) to (iii) and comparison with the previous approach for a single heat bath

For an arbitrary initial system-bath state $\rho_{SB}(0)$ the change in observational entropy can be expressed as

$$\Delta S_{\text{obs}} = \Delta S_{\text{Sh}}[p_s(t)] + \Delta S_{\text{obs}}^{E_B} - \Delta I[p_s(t)]$$

$$= S_{\text{Sh}}[p_s(t)] - S_{\text{Sh}}[p_s(0)] + S_{\text{vN}} \left[ \sum_E p_E(t) \rho_B(E, t) \right] - S_{\text{vN}} \left[ \sum_E p_E(0) \rho_B(E, 0) \right]$$

$$+ \sum_E p_E(t) D[\rho_B(E, t) \parallel \rho_{\text{mic}}(E)] - \sum_E p_E(0) D[\rho_B(E, 0) \parallel \rho_{\text{mic}}(E)] - I_{s;E}(t) + I_{s;E}(0), \quad (20)$$
where we used Eqs. (10) and (12). Here, \( p_s(t) = \langle s | \rho_S(t) | s \rangle \) and \( \rho_B(E,t) = \Pi_{E \rho B}(t) \Pi_E / \rho_E(t) \). Next, we assume the initial state and width \( \delta \) to be as described in Sec. II B, such that \( S_{SB}^E(0) = S_{\gamma N}(\pi_B) \). This allows us to confirm

\[
\Delta S_{\text{obs}} = S_{SB}[p_s(t)] - S_{SB}[p_s(0)] + S_{\gamma N} \left[ \sum_E p_E(t) \rho_B(E,t) \right] - S_{\gamma N}(\pi_B) \\
+ \sum_E p_E(t) D[\rho_B(E,t) \| \rho_{\text{mic}}(E)] - I[p_sE(t)].
\] (21)

Now, as the entropy is preserved during any unitary evolution, we get \( S_{SB}[p_s(0)] + S_{\gamma N}(\pi_B) = S_{\gamma N}[\rho_{SB}(t)] \) where \( \rho_{SB}(t) \) is the time-evolved state starting from the initial state \( \rho_{SB}(0) = \sum_s p_s(0) | s \rangle \langle s | \otimes \pi_B \). Writing also \( S_{\gamma N}[\rho_{SB}(t)] = S_{\gamma N}[\rho_S(t)] + S_{\gamma N}[\rho_B(t)] - I[\rho_{SB}(t)] \), we get

\[
\Delta S_{\text{obs}} = S_{SB}[p_s(t)] - S_{\gamma N}[\rho_S(t)] + S_{\gamma N} \left[ \sum_E p_E(t) \rho_B(E,t) \right] - S_{\gamma N}[\rho_B(t)] \\
+ \sum_E p_E(t) D[\rho_B(E,t) \| \rho_{\text{mic}}(E)] + I[\rho_{SB}(t)] - I[p_sE(t)].
\] (22)

The positivity of \( \Delta S_{\text{obs}} \) is now evident. First, by using that a projective measurement increases the entropy on average, Theorem 11.9 in Ref. [43], we confirm that

\[
S_{SB}[p_s(t)] \geq S_{\gamma N}[\rho_S(t)], \quad S_{\gamma N} \left[ \sum_E p_E(t) \rho_B(E,t) \right] \geq S_{\gamma N}[\rho_B(t)].
\] (23)

We expect both contributions, however, to be rather small. First, the change in system entropy due to the measurement is at most \( \ln \text{dim}(\mathcal{H}_S) \) and will likely be much smaller. Especially, we are free to choose the basis of the final system measurement at time \( t \) such that we can let it coincide with the eigenbasis of \( \rho_S(t) \). Second, a large change in bath entropy due to the final measurement requires the existence of large coherences \( \text{tr}_B \{ \Pi_E \rho_B(t) \Pi_{E'} \} \neq 0 \) between different energy sectors \( E \neq E' \). This also seems very unlikely as it would imply the existence of macroscopic Schrödinger cat states in the bath. Finally, another small contribution comes from the difference in mutual information, which always obeys

\[
I[\rho_{SB}(t)] = D[\rho_{SB}(t) \| \rho_S(t) \otimes \rho_B(t)] \\
\geq D \left[ \sum_{s,E} p_sE(t) | s \rangle \langle s | \otimes \rho_{\text{mic}}(E) \right] \left[ \sum_s p_s(t) | s \rangle \langle s | \otimes \sum_E p_E(t) \rho_{\text{mic}}(E) \right] = I[p_sE(t)],
\] (24)

This follows from monotonicity of relative entropy [44, 45] by noting that \( \sum_{s,E} p_sE(t) | s \rangle \langle s | \otimes \rho_{\text{mic}}(E) = \Phi \rho_{SB}(t) \) with the completely positive and trace-preserving map defined via

\[
\Phi \rho_{SB} = \sum_E \text{tr}_B \{ | s \rangle \langle s | \Pi_E \rho_{SB} | s \rangle \langle s | \Pi_E \} \otimes \rho_{\text{mic}}(E).
\] (25)

Thus, all together we can conclude that

\[
\Delta S_{\text{obs}} \gtrsim \sum_E p_E(t) D[\rho_B(E,t) \| \rho_{\text{mic}}(E)] \geq 0.
\] (26)

where we used the symbol “\( \gtrsim \)" to indicate that the difference between the two sites of the inequality is expected to be rather small in the typical situation of a large heat bath. The only part, which can scale extensively with time, is \( \sum_E p_E(t) D[\rho_B(E,t) \| \rho_{\text{mic}}(E)] \). Therefore, we conclude that \( \text{entropy production arises because for a given energy } E \text{ of the bath we lose track of its exact microstate} \) compared to the maximal uninformative microcanonical ensemble, where all microstates are assumed to be equally likely. Thus, we have confirmed the points (i) and (ii) from above.

Next, to confirm (iii), we want to show that for a weakly coupled, macroscopic bath the change in observational bath entropy is proportional to its change in energy. For that purpose it is indeed crucial to assume that the bath is initially in a Gibbs state. Then, we write \( p_E(t) = \pi_E + \epsilon q_E(t) \) with \( \pi_E = e^{-\beta E V_B / Z_B} \) and \( q_E(t) \) is a set of numbers such that \( \sum_E q_E(t) = 0 \). Now, our assumption is that \( \epsilon \) is a small parameter, i.e., the distribution of energies in the bath remains close to the canonical probabilities throughout the time-evolution. This should be typically justified for
a weakly coupled, macroscopic bath. Notice that this does not imply that the entire bath state \( \rho_B(t) \) is close to the Gibbs state \( \pi_B \). Then, we can write

\[
\Delta S_{\text{obs}}^E = - \sum_E [\pi_E + \epsilon q_E(t)] \ln \frac{\pi_E + \epsilon q_E(t)}{V_E} + \sum_E \pi_E \ln \frac{\pi_E}{V_E} = \beta \Delta E_B + \mathcal{O}(\epsilon^2)
\]

with the change in bath energy \( \Delta E_B = \epsilon \sum_E E q_E(t) \). Hence,

\[
\Delta S_{\text{obs}} = \Delta S_{\text{Sh}}[\rho_S(t)] + \Delta S_{\text{obs}}^E - I[p_{sE}(t)] \approx \Delta S_{\text{Sh}}[\rho_S(t)] - \beta Q \geq 0,
\]

where we ignored the small contribution \( I[p_{sE}(t)] \) at the end. We have also identified \( Q = -\Delta E_B \), which is justified in the limit considered here to derive \( \Delta S_{\text{obs}}^E \approx \beta \Delta E_B \).

Thus, as a preliminary conclusion, we have shown that \( \Sigma \equiv \Delta S_{\text{obs}} \) fulfills the three desired properties (i) to (iii) and therefore, provides a more suitable candidate for entropy production than \( \tilde{\Sigma} \) from Eq. (5) because the latter cannot be expressed as the change of a meaningful thermodynamic entropy for the system and the bath.

Nevertheless, it is instructive to compare \( \Sigma \) and \( \tilde{\Sigma} \) quantitatively. From Eqs. (22) and (5) we obtain

\[
\Sigma = \tilde{\Sigma} = S_{\text{Sh}}[p_s(t)] + S_{\text{VN}} \left[ \sum_E p_E(t) \rho_B(E,t) \right] - S_{\text{VN}}[\rho_{SB}(t)] + \sum_E p_E(t) D[\rho_B(E,t)\|\rho_{\text{mic}}(E)] - I[p_{sE}(t)]
\]

where we also made use of Eq. (13). Thus, we end up with the compact expression

\[
\Sigma - \tilde{\Sigma} = S_{\text{Sh}}[p_s(t)] - S_{\text{VN}}[\rho_{SB}(t)] - D[\rho_B(t)\|\pi_B(t)] - I[p_{sE}(t)].
\]

By combining this result with the second and forth term of Eq. (29), we verify

\[
S_{\text{VN}} \left[ \sum_E p_E(t) \rho_B(E,t) \right] + \sum_E p_E(t) D[\rho_B(E,t)\|\rho_{\text{mic}}(E)] - D[\rho_B(t)\|\pi_B] = S_{\text{VN}}[\rho_B(t)] - D[p_E(t)\|\pi_B(E)],
\]

where we also made use of Eq. (30). Thus, we obtain

\[
\Sigma = \tilde{\Sigma} = S_{\text{Sh}}[p_s(t)] - S_{\text{VN}}[\rho_{SB}(t)] - D[p_E(t)\|\pi_B(E)] - I[p_{sE}(t)].
\]

Taken together, the first two terms are non-negative (and identical to zero for classical systems), whereas each of the remaining two terms is negative. In the weak coupling regime investigated above, we expect all terms to be rather small. In fact, if \( p_E(t) = \pi_E + \epsilon q_E(t) \) as above, then \( D[p_E(t)\|\pi_B(E)] = \mathcal{O}(\epsilon^2) \) such that we can conclude \( \Sigma \approx \tilde{\Sigma} \). Outside this regime, the only possibly unbounded term is \( D[p_E(t)\|\pi_B(E)] \) as the number of populated energy levels for a very small \( \delta \) can become very large. We therefore expect that, typically, we have

\[
\tilde{\Sigma} \geq \Sigma \geq 0.
\]

This means that the previous approach typically yields a larger entropy production than our novel definition. Intuitively, this makes sense: in our approach we ideally know the entire distribution of energies in the bath whereas in the previous approach only knowing the average energy flow to the bath is sufficient. Clearly, our approach stores more information and hence, less entropy is produced.

### D. Extension to multiple heat baths

The extension to multiple heat baths labeled by \( \nu \in \{1, \ldots, N\} \) is straightforward by measuring the energy of each bath. The observational entropy is in this case

\[
S_{\text{obs}} = - \sum_{s,E_1,\ldots,E_N} p_{sE_1\ldots E_N} \ln \frac{p_{sE_1\ldots E_N}}{V_E_1 \ldots V_E_N}. \tag{34}
\]
Here, we used that the number of microstates naturally factorizes, \( V_{E_1 \ldots E_N} = \text{tr}_{B_1 \ldots B_N} \{ \Pi_{E_1} \ldots \Pi_{E_N} \} = V_{E_1} \ldots V_{E_N} \), where \( \Pi_{E} \) describes the projector associated to measurement outcome \( E_{\nu} \) of bath \( \nu \). Furthermore, the natural generalization of the initial state (3) to multiple baths is

\[
\rho_{\text{tot}}(0) = \rho_S(0) \otimes \pi_{B_1} \otimes \cdots \otimes \pi_{B_N},
\]

which was also used in Ref. [9]. We note that every bath can have initially a different inverse temperature, i.e.,

\[
\pi_{B_\nu} = e^{-\beta_\nu H_{B_\nu}} / Z_{B_\nu}.
\]

Under these circumstances (assuming that \( \delta \) is chosen as in Sec. II B) we easily confirm that the initial observational entropy is identical to

\[
S_{\text{obs}}(0) = S_{vN} \left[ \sum_s \rho_s(0) |s\rangle \langle s| \otimes \pi_{B_1} \otimes \cdots \otimes \pi_{B_N} \right] = S_{vN} [\rho_{\text{tot}}(t)].
\]

Furthermore, the observational entropy at time \( t \) can be split into its ‘local’ parts and its correlations, similar to Eq. (10). Specifically,

\[
S_{\text{obs}}(t) = - \sum_s \rho_s(t) \ln \rho_s(t) - \sum_{\nu} \sum_{E_{\nu}} p_{E_{\nu}}(t) \ln \frac{p_{E_{\nu}}(t)}{V_{E_{\nu}}} - \sum_{s,E_{1},\ldots,E_{N}} \rho_{sE_{1} \ldots E_{N}}(t) \ln \frac{\rho_{sE_{1} \ldots E_{N}}(t)}{p_s(t)p_{E_{1}}(t) \ldots p_{E_{N}}(t)}
\]

\[
= S_{\text{Sh}}[\rho_s(t)] + \sum_{\nu} S^{E_{\nu}}_{\text{obs}}(t) - I_{\text{cor}}[\rho_{sE_{1} \ldots E_{N}}(t)].
\]

Further use of relation (12) reveals that

\[
S_{\text{obs}}(t) = S_{\text{Sh}}[\rho_S(t)] + \sum_{\nu} S_{vN} \left[ \sum_{E_{\nu}} p_{E_{\nu}} \rho_{B_{\nu}}(E_{\nu}, t) \right] + \sum_{\nu} \sum_{E_{\nu}} p_{E_{\nu}} D[\rho_B(E_{\nu}, t) \| \rho_{\text{mic}}(E_{\nu})] - I_{\text{cor}}[\rho_{sE_{1} \ldots E_{N}}(t)].
\]

Thus, similarly to Eq. (22), the change in observational entropy can be split into a family of terms, whose non-negativity is evident:

\[
\Delta S_{\text{obs}} = S_{\text{Sh}}[\rho_S(t)] - S_{vN}[\rho_S(t)] + \sum_{\nu} \left\{ S_{vN} \left[ \sum_{E_{\nu}} p_{E_{\nu}} \rho_{B_{\nu}}(E_{\nu}, t) \right] - S_{vN}[\rho_{B_{\nu}}(t)] \right\}
\]

\[
+ \sum_{\nu} \sum_{E_{\nu}} p_{E_{\nu}} D[\rho_B(E_{\nu}, t) \| \rho_{\text{mic}}(E_{\nu})] + I_{\text{cor}}[\rho_{\text{tot}}(t)] - I_{\text{cor}}[\rho_{sE_{1} \ldots E_{N}}(t)].
\]

As before, we expect the non-negativity of the first line to be rather small (especially, it is exactly zero for classical systems) such that

\[
\Delta S_{\text{obs}} \geq \sum_{\nu} \sum_{E_{\nu}} p_{E_{\nu}} D[\rho_B(E_{\nu}, t) \| \rho_{\text{mic}}(E_{\nu})] + I_{\text{cor}}[\rho_{\text{tot}}(t)] - I_{\text{cor}}[\rho_{sE_{1} \ldots E_{N}}(t)].
\]

Here, we introduced the notation

\[
I_{\text{cor}}[\rho_{\text{tot}}(t)] = S_{vN}[\rho_S(t)] + \sum_{\nu} S_{vN}[\rho_{B_{\nu}}(t)] - S_{vN}[\rho_{\text{tot}}(t)] = D[\rho_{\text{tot}}(t) \| \rho_S(t) \otimes \rho_{B_1}(t) \otimes \cdots \otimes \rho_{B_N}(t)].
\]

Since we can also write

\[
I_{\text{cor}}[\rho_{sE_{1} \ldots E_{N}}(t)] =
\]

\[
D \left[ \sum_{s,E_{1},\ldots,E_{N}} p_{sE_{1} \ldots E_{N}}(t) |s\rangle \langle s| \rho_{\text{mic}}(E_{1}) \cdots \rho_{\text{mic}}(E_{N}) \left| \sum_s p_s(t) |s\rangle \langle s| \sum_{E_{1}} p_{E_{1}}(t) \rho_{\text{mic}}(E_{1}) \cdots \sum_{E_{N}} p_{E_{N}}(t) \rho_{\text{mic}}(E_{N}) \right] .
\]

we can confirm \( I_{\text{cor}}[\rho_{\text{tot}}(t)] - I_{\text{cor}}[\rho_{sE_{1} \ldots E_{N}}(t)] \geq 0 \) similar to Eq. (24). However, in contrast to the case of a single heat bath, we can no longer expect this contribution to be small as the different baths can become correlated. This is especially true outside the weak coupling case. Therefore, in the case of multiple baths we expect that there are two major contributions to the entropy production. In any case, \( \Delta S_{\text{obs}} \geq 0 \) is ensured.
Furthermore, following the same procedure as above, we can confirm in the weak coupling case that $\Delta S_{\text{obs}}^B = \beta_\nu \Delta E_B$. In addition, we expect in the weak coupling limit that $I_{\text{cor}}[p_{s,t}(t)] \gtrsim I_{\text{cor}}[p_{s,E_1} \cdots E_N(t)] \approx \sum_\nu I[p_{s,E_\nu}(t)]$, i.e., each bath acts like a separate bath entering independently the master equation describing the system \cite{6,7,42}.\footnote{A critical discussion of this point can be found in Ref. \cite{46}.} Hence, we can write $\Delta S_{\text{obs}}(t) \approx \Delta S_{\text{Sh}}[p_{s}(t)] - \sum_\nu \beta_\nu Q_\nu \geq 0$. This provides a microscopic derivation of the phenomenological second law of nonequilibrium thermodynamics \cite{2}.

\section*{E. Additional observations}

We end this general section with a couple of interesting observations:

\textbf{Observation 1.} The crucial ingredient to prove point (i), positivity of $\Delta S_{\text{obs}}$, is that Eq. \cite{14} vanishes. This is not only true for a Gibbs state and a small enough measurement width $\delta$. Indeed, Eq. \cite{14} can be zero for many different initial energy distributions $p_E(0)$ as long as the distribution of microstates within a given energy window is very close to the microcanonical ensemble. The initial state of the bath can even contain quantum coherences between different energy sectors as those get killed during the initial measurement.

\textbf{Observation 2.} One can also choose different measurements of the bath and positivity of $\Delta S_{\text{obs}}$ will still hold as long as the initial observational entropy of the bath coincides with the von Neumann entropy of the average post measurement state. In that respect the energy only seems to be an outstanding observable due to its connection to the first law. To capture the effect of multiple conserved quantities, we can consider additional measurements, e.g., of the energy and particle number of the bath in case of a grand-canonical reservoir.

\textbf{Observation 3.} We also do not expect the initial product state assumption to be crucial. An initially correlated system-bath state lowers the entropy production by at most $I[p_{s,E}(0)]$, which is typically negligible with respect to the positive terms appearing in Eq. \cite{26}.

\textbf{Observation 4.} Instead of taking into account correlations between the measurement results of the system state and the bath energy, we could also neglect them in the definition of observational entropy. All the three points (i) to (iii) would remain valid for the choice $S_{\text{obs}}(t) \equiv S_{\text{Sh}}[p_s(t)] + S_{\text{obs}}^E(t)$. In particular, we would typically have $\Delta S_{\text{obs}} \approx \Delta S_{\text{obs}}$.

\textbf{Observation 5.} Finally, we emphasize that none of our results depends on the particular form of the Hamiltonian. Especially, the system-bath coupling could be time-dependent, $H_{SB} = H_{SB}(\lambda_t)$, and even the bath Hamiltonian could depend on time, $H_B = H_B(\lambda_t)$.

\section{III. THE MICROCANONICAL MASTER EQUATION}

In this section we illustrate our general findings in the limit of a weakly coupled, Markovian bath. In contrast to conventional master equations \cite{5-8}, we will derive a master equation describing the evolution of the system state and the bath energies by using a correlated projection-operator method. Such a master equation was first derived by Esposito and Gaspard \cite{47} and we repeat a (slightly more generalized) derivation in Appendix A. Here, we will investigate in detail the analytical and thermodynamic properties of this master equation, which was not done in Ref. \cite{47}. We will call this approach the ‘microcanonical master equation’ (MME) in the following.

The MME is a Pauli-like rate master equation for the probabilities $p_{s,E}(t)$ to find the system in state $s$ and the energy of the bath at $E$ at time $t$. It reads

$$
\dot{p}_{s,E}(t) = \sum_{\alpha,\gamma} \sum_{s'} \frac{2\pi}{V_E} \mathcal{R}\{S^s_{s'} S^{s'}_{\gamma} f_{\alpha \gamma}(E,E + \epsilon_s - \epsilon_{s'})\} \left[ \frac{V_E}{V_E + \epsilon_s - \epsilon_{s'}} p_{s',E + \epsilon_s -\epsilon_{s'}(t)} - p_{s,E}(t) \right].
$$

Here, the overall timescale of the dynamics is governed by the rate $\mathcal{R}\{S^s_{s'} S^{s'}_{\gamma} f_{\alpha \gamma}(E,E + \epsilon_s - \epsilon_{s'})\}$. If one assumes that the system-bath coupling Hamiltonian reads $H_{SB} = \sum_\alpha S_\alpha \otimes B_\alpha$, where $S_\alpha (B_\alpha)$ are Hermitian system (bath) operators, then $S^{s'}_{s'} = (s|S_\alpha|s')$ describes the transition matrix elements with respect to the basis $|s\rangle$, which is assumed in this section to be the (non-degenerate) energy eigenbasis of $H_S$. Furthermore, the function $f_{\alpha \gamma}(E,E') \equiv \text{tr}_B\{\Pi_E B_\alpha \Pi_{E'} B_\gamma\}$ describes how well the energies in the bath get redistributed. In obyes the useful relations \cite{A6}.

We remark that Eq. \cite{43} reduces to Eq. \cite{42} of Ref. \cite{47} in the limit of a single system coupling operator $S_\alpha = \delta_\alpha 1 S$. Multiple baths can be easily included by summing over $\nu$ and adding this superscript to $S_\alpha$, $f$ and $V$, but we will only consider a single bath here. As this equation describes the time-evolution of all energies, we will indeed find out below that the dynamics of this equation are entropy dominated. Furthermore, we remark that driven system
energies can be considered by replacing $\epsilon_s$ by $\epsilon_s(\lambda_t)$ provided that the change of energies is slow compared to the decay of the bath correlation functions.

A. Properties

1. Reduction to the conventional Pauli master equation

As a simple crosscheck we investigate the limit in which our MME reduces to the conventional Pauli master equation derived within the Born-Markov-secular approximation [8]. Formally, we can write Eq. (43) after summing over $E$ as

$$\partial_t p_s(t) = \sum_E \sum_{\alpha, \gamma} \sum_{s', s} \frac{2\pi}{V_E} \mathfrak{R}\{S_{\alpha}^{s,s'} S_{\gamma}^{s} f_{\alpha \gamma}(E, E + \epsilon_s - \epsilon_{s'})\} \left[ \frac{V_E}{V_{E + \epsilon_s - \epsilon_{s'}} - \epsilon_{s'}} p_{E + \epsilon_s - \epsilon_{s'}|s'}(t)p_{s'}(t) - p_{E|s}(t)p_s(t) \right].$$

Here, we have introduced the conditional probability $p_{E|s}(t) \equiv p_{sE}(t)/p_s(t)$ and we will now assume that this is approximately given by $p_{E|s}(t) \approx V_E e^{-\beta E}/Z_B$ for all $s$ and all times $t$. This simplifies the expression to

$$\partial_t p_s(t) = \frac{2\pi}{Z_B} \sum_{E} \sum_{\alpha, \gamma} \sum_{s', s} \mathfrak{R}\{S_{\alpha}^{s,s'} S_{\gamma}^{s} f_{\alpha \gamma}(E, E + \epsilon_s - \epsilon_{s'})\} \left[ e^{-\beta(E + \epsilon_s - \epsilon_{s'})} p_{s'}(t) - e^{-\beta E} p_s(t) \right].$$

We now introduce the functions $g_{\alpha \gamma}(\epsilon_s - \epsilon_{s'}) \equiv \sum_E f_{\alpha \gamma}(E, E + \epsilon_s - \epsilon_{s'}) e^{-\beta E}$, which obey the symmetries $g_{\alpha \gamma}(\epsilon_s - \epsilon_{s'}) = e^{\beta(\epsilon_s - \epsilon_{s'})} g_{\gamma \alpha}(\epsilon_s - \epsilon_{s'})$ and $g_{\alpha \gamma}^*(\epsilon_s - \epsilon_{s'}) = g_{\gamma \alpha}(\epsilon_s - \epsilon_{s'})$. They allow us to write

$$\partial_t p_s(t) = 2\pi \sum_{s, \alpha, \gamma} \sum_{s'} \mathfrak{R}\{S_{\alpha}^{ss'} S_{\gamma}^{s} g_{\alpha \gamma}(\epsilon_s - \epsilon_{s'})\} \left[ e^{\beta(\epsilon_{s'} - \epsilon_s)} p_{s'}(t) - p_s(t) \right].$$

This corresponds to the typical Pauli master equation [8] with the rates satisfying local detailed balance, i.e., the rate to jump from $s'$ to $s$ is enhanced by a factor $e^{\beta(\epsilon_{s'} - \epsilon_s)}$ compared to the inverse jump rate from $s$ to $s'$ if $\epsilon_{s'} > \epsilon_s$.

2. Conservation of energy

To confirm conservation of energy, we note that

$$\sum_{s,E} \sum_{\alpha, \gamma} \sum_{s'} (\epsilon_s + E) \mathfrak{R}\{S_{\alpha}^{ss'} S_{\gamma}^{s} f_{\alpha \gamma}(E, E + \epsilon_s - \epsilon_{s'})\} \frac{1}{V_{E + \epsilon_s - \epsilon_{s'}}} p_{s', E + \epsilon_s - \epsilon_{s'}}(t)$$

$$= \sum_{s', E} \sum_{\alpha, \gamma} \sum_{s'} (\epsilon_{s'} + E') \mathfrak{R}\{S_{\alpha}^{s's} S_{\gamma}^{s} f_{\alpha \gamma}(E' - \epsilon_s + \epsilon_{s'}, E')\} \frac{1}{V_{E'}} p_{s', E'}(t)$$

$$= \sum_{s, \alpha, \gamma} \sum_{s'} (\epsilon_s + E) \mathfrak{R}\{S_{\alpha}^{ss'} S_{\gamma}^{s} f_{\alpha \gamma}(E, E + \epsilon_s - \epsilon_{s'})\} \frac{1}{V_E} p_{sE}(t),$$

where we used the symmetry relation (A6) and made use of the freedom to relabel indices within the summation. Equation (47) holds for any fixed time and even in presence of driving when $\epsilon_s = \epsilon_s(\lambda_t)$. In this case one confirms that

$$\frac{d}{dt} [\Delta E_S(t) + \Delta E_B(t)] = \frac{d}{dt} \sum_{s, E} [\epsilon_s(\lambda_t) + E] p_{sE}(t) = \sum_s \dot{\epsilon}_s(\lambda_t) p_s(t) \equiv \dot{W}.$$

This is the first law of thermodynamics in presence of driving.

Interestingly, for an undriven system Eq. (47) even implies that

$$\frac{d}{dt} (f(\epsilon_s + E)) \equiv \frac{d}{dt} \sum_{s, E} f(\epsilon_s + E) p_{sE}(t) = 0$$

for an arbitrary function $f(E_{\text{tot}})$ of the total energy $E_{\text{tot}} = \epsilon_s + E$. We can call this strict energy conservation. It essentially implies that there is only one random variable in the problem (and not the two $\epsilon_s$ and $E$) because the distribution for $E_{\text{tot}}$ remains fixed for all times. This conclusion holds, however, only in absence of driving.
3. Steady state of the Pauli MME

We here consider the case where \( \epsilon_s \) is held fixed in time (i.e., \( \dot{\lambda}_s = 0 \)) and we ask for which state \( \bar{p}_{s,E} \) the Pauli MME (43) evaluates to zero. We call \( \bar{p}_{s,E} \) a steady state in this case. One point we can immediately recognize from Eq. (43) is that every state \( \bar{p}_{s,E} \) which fulfills

\[
\frac{V_E}{V_{E+\epsilon_s-\epsilon_{s'}}} = \frac{\bar{p}(s,E)}{\bar{p}(s',E+\epsilon_s-\epsilon_{s'})}
\]

is a steady state. However, due to the fact that the energy is strictly conserved, there are infinitely many possible steady states; indeed even infinitely many for every initial energy \( \langle \epsilon_s + E \rangle(0) = E_0 \) depending on how the probabilities are initially distributed. For instance, one possible steady state is the overall Gibbs state

\[
\bar{p}_{s,E} = \frac{e^{-\beta\epsilon_s} V_E e^{-\beta E}}{Z_S} \equiv \pi_{s,E},
\]

where \( \beta \) must be fixed through \( E_0 = -\partial \beta \ln(Z_S Z_B) \).

On the other hand, imagine that we start with a definite initial condition such as \( p_{s,E}(t = 0) = \delta_{s,0} \delta_{E,E_0} \) and we assume that the energy eigenvalues are ordered according to \( \epsilon_n > \cdots > \epsilon_1 > \epsilon_0 \equiv 0 \). Then, the dynamics are restricted to the following states with energies \( (\epsilon_s, E) \):

\[
(0, E_0), (\epsilon_1, E_0 - \epsilon_1), \ldots, (\epsilon_{n-1}, E_0 - \epsilon_{n-1}), (\epsilon_n, E_0 - \epsilon_n).
\]

Note that the dynamics is not restricted to jumps between nearest neighbours as one might be tempted to think.\(^4\) The ratio of the rates to jump from one state to another are

\[
\frac{\text{rate}[(\epsilon_s, E_0 - \epsilon_s) \rightarrow (\epsilon_{s'}, E_0 - \epsilon_{s'})]}{\text{rate}[(\epsilon_{s'}, E_0 - \epsilon_{s'}) \rightarrow (\epsilon_s, E_0 - \epsilon_s)]} = \frac{V_{E_0-\epsilon_{s'}}}{V_{E_0-\epsilon_s}},
\]

which can be interpreted as a purely entropic factor. Thus, the dynamics of the MME are entropy dominated. Typically, one expects that \( V_{E'} < V_E \) if \( E' > E \). Then, the system tends to prefer low energies in order to increase the entropy of the environment. A particularly interesting case arises if the bath behaves like an ideal heat bath. Using Boltzmann’s entropy formula, we infer that \( V_E = e^{S_{\text{Sh}}(E)/k_B} \) where \( S_B(E) \) is the entropy of the bath at energy \( E \). Now, the assumption of an ideal heat bath enters by invoking the standard definition of temperature, \( T^{-1} = S_B'(E) \), which allows us to derive

\[
\frac{V_{E_0-\epsilon_{s'}}}{V_{E_0-\epsilon_{s}}} = e^{\beta(\epsilon_{s'}-\epsilon_s)}. \]

This implies that the ratio of the rates (53) fulfills the conventional local detailed balance relation. One steady state of the MME with initial condition \( p_{s,E}(t = 0) = \delta_{s,0} \delta_{E,E_0} \) is then given by

\[
\bar{p}_{s,E} = \begin{cases} 
\frac{e^{-\beta\epsilon_s}}{Z_S} & \text{if } E = E_0 - \epsilon_s \\
0 & \text{otherwise}
\end{cases}
\]

That is to say, the system equilibrated to the canonical ensemble with the temperature imposed by the initial energy of the bath which becomes equally distributed over the available phase space. This is nothing else than the equivalence of ensembles, i.e., the reduced state of a weakly coupled subsystem is a canonical distribution if the entire system has a fixed energy \( E_0 \). Note that the above state indeed fulfills \( \sum_{s,E}(\epsilon_s + E)\bar{p}_{s,E} = E_0 \).

B. Entropy production and observational entropy

Conventionally, the entropy production for the Pauli master equation (46) can be expressed as

\[
\dot{S}(t) = \frac{d}{dt} S_{\text{Sh}}[p_s(t)] - \beta \dot{Q}(t) = -\frac{\partial}{\partial t} D[p_s(t)]\|\pi_s(\lambda_t)] \geq 0,
\]

where \( \pi_s(\lambda_t) = e^{-\beta \epsilon_s(\lambda_t)}/Z_S(\lambda_t) \) is the instantaneous canonical equilibrium state of the system at time \( t \). The derivative in Eq. (55) is evaluated with respect to a fixed \( \lambda_t \) and positivity of the entropy production follows from

\[^4\] The precise topology of the network depends strongly on the prefactor \( \Re\{S_{\alpha}^{s,s'} S_{\alpha}^{s'} f_{\alpha\gamma}(E, E + \epsilon_s - \epsilon_{s'})\} \).
the two facts that the dynamics are Markovian and that \( \pi_s(\lambda_t) \) is an instantaneous steady state of the dynamics, see, e.g., Refs. [6, 8, 26]. Finally, \( \dot{Q}(t) = \sum_s \epsilon_s(\lambda_t) \partial_t p_s(t) \) is the heat flow into the system.

Similarly, also for the MME we can derive an always positive entropy production rate by considering

\[
\dot{\Sigma}(t) \equiv - \frac{\partial}{\partial t} \bigg|_{\lambda_t} D[\bar{p}_{sE}(t)||\bar{p}_{sE}(\lambda_t)] \geq 0,
\]  

(56)

where \( \bar{p}_{sE}(\lambda_t) \) is any admissible steady state of the Pauli MME (independent of the initial condition), which is allowed to depend parametrically on time through \( \lambda_t \). As we have a multitude of possible steady states, see Sec. III A 3, there are many different possible choices, each leading to a different positive ‘entropy production’ rate. The choice, which leads to the desired final result, turns out to be the Gibbs state from Eq. (51), where \( \epsilon_s = \epsilon_s(\lambda_t) \) is allowed to be time-dependent. Indeed, upon integration of Eq. (56) we have

\[
\Sigma(t) = \int_0^t ds \begin{pmatrix} -\frac{d}{ds} + \lambda_t \frac{\partial}{\partial \lambda_t} \end{pmatrix} D[\bar{p}_{sE}(s)||\bar{p}_{sE}(\lambda_s)]
= -D[\bar{p}_{sE}(t)||\bar{p}_{sE}(\lambda_t)] + D[\bar{p}_{sE}(0)||\bar{p}_{sE}(\lambda_0)] - \int_0^t ds \dot{\lambda}_t \frac{\partial}{\partial \lambda_t} \sum_{s,E} p_{sE}(s) \ln \pi_{sE}(\lambda_s),
\]  

(57)

where we used the chain rule \( \frac{d}{dt} = \frac{\partial}{\partial \lambda_t} |_{\lambda_t} + \dot{\lambda}_t \frac{\partial}{\partial \lambda_t} \). The particular form of \( \pi_{sE}(\lambda_t) \) reveals after some straightforward manipulations that

\[
\Sigma(t) = - \sum_{s,E} \bar{p}_{sE}(t) \ln \frac{\bar{p}_{sE}(t)}{V_E} + \sum_{s,E} \bar{p}_{sE}(0) \ln \frac{\bar{p}_{sE}(0)}{V_E}
+ \beta \sum_{s,E} \left\{ -[\epsilon_s(\lambda_t) + E] \bar{p}_{sE}(t) + [\epsilon_s(\lambda_0) + E] \bar{p}_{sE}(0) + \int_0^t ds \epsilon_s(\lambda_s) \bar{p}_{sE}(s) \right\}
= \Delta \Sigma_{\text{obs}}.
\]

Here, for the final step we used the first law (48). Thus, we have confirmed that \( \Sigma(t) = \Delta \Sigma_{\text{obs}} \geq 0 \) also follows directly from the Pauli MME.

Finally, we compare \( \Sigma(t) \) with \( \Sigma(t) \) obtained by integrating Eq. (55). First, we use that we can express the heat flow within the MME approach as

\[
- \beta Q(t) = \beta \sum_E [E \bar{p}_E(t) - p_E(0)] = \sum_E [p_E(t) - p_E(0)] \ln V_E + \Delta \Sigma_{\text{obs}}[p_E(t)] + D[p_E(t)||\pi_E],
\]

(59)

if we assume that \( p_E(0) = \pi_E \). If the initial state \( p_{sE}(0) = p_s(0)p_E(0) \) is furthermore decorrelated, we can confirm that

\[
\Sigma(t) - \Sigma(t) = -I[p_{sE}(t)] - D[p_E(t)||\pi_E] \leq 0.
\]

(60)

This result is in direct analogy to our general finding (32). Thus, we have re-derived all our general findings within the particularly important limit of a system weakly coupled to a Markovian bath. Notice that we could even derive a stronger statement in that limit, namely that the rate of entropy production (56) is positive. This is usually not the case within the general setup of Sec. II. Conditions which ensure the positivity of the entropy production rate \( \frac{d}{dt} \Sigma_{\text{obs}}(t) \) are missing; within the standard approach reviewed in Sec. 1B answers were partially found in Refs. [26, 48].

IV. CONCLUSIONS

We have put forward a novel approach to understand and quantify entropy production in open (quantum) systems driven arbitrarily far from equilibrium. For this purpose we constructed a suitable notion of entropy for the entire universe (system plus bath) based on the recently introduced observational entropy from Refs. [1–3]. Then, using very similar steps as in previous approaches (conservation of global von Neumann entropy, special – but slightly more general – form of the initial system-bath state, see Sec. II B), we showed that the change in observational entropy is always positive for arbitrary dynamics, has a clear information-theoretic interpretation, and can be linked to the standard expression (2) in the limit of weak system-bath coupling. Therefore, our novel notion fulfills the three
minimum requirements (i) to (iii), but – moreover and most importantly – it fulfills them expressed as a change in a thermodynamically meaningful definition of global entropy. Thus, we were able to microscopically derive the statement that \textit{entropy production measures the change in thermodynamic entropy of the universe, which is always positive.}

Quite interestingly, in the conventionally considered weak coupling limit we showed that the quantitative difference between our and the former approach is negligible. This reassures the consistency of both, our and the former approach. Outside the weak coupling regime, interesting difference could appear. As we have also stated the condition which needs to be fulfilled to test this theory experimentally [Eq. (19)], we have the hope that it will be possible to measure the global entropy production in the future, for instance, in cold atoms [49] or in electronic nanostructures coupled to mesoscopic heat baths [50].

Finally, we believe that our results provide strong evidence that observational entropy as advertised in Refs. [1–3] provides a good candidate for thermodynamic entropy of isolated out-of-equilibrium systems.

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We start with the standard system-bath Hamiltonian $H_{\text{tot}} = H_S + H_{SB} + H_B$ as usual and assume that the interaction can be decomposed as $H_{SB} = \sum \alpha S_{\alpha} \otimes B_{\alpha}$, where $S_{\alpha}$ ($B_{\alpha}$) are Hermitian system (bath) operators. The standard time-convolutionless Nakajima-Zwanzig projection operator method predicts within the weak coupling and the interaction picture that [8]

$$\partial_t \mathcal{P} \hat{\rho}(t) = \int_0^t ds \mathcal{P} \mathcal{L}_1(t) \mathcal{L}_1(s) \mathcal{P} \hat{\rho}(t),$$

(A1)
where \( L_I(t) \rho = -i[H_{SB}, \rho] \). As our projection superoperator we choose

\[
P \rho = \sum_E \text{tr}_B \{ \Pi_E \rho \} \otimes \rho_{\text{mic}}(E).
\]

(A2)

Below, we will denote \( \rho_S(E, t) \equiv \text{tr}_B \{ P \rho(t) \} = \text{tr}_B \{ \Pi_E \rho \} \). We remark that the validity of Eq. (A1) is only ensured if \((1 - P) \rho(0) = 0\). Furthermore, we assume that \( \text{tr}_B \{ B_\alpha \rho_{\text{mic}}(E) \} = 0 \) for all \( \alpha \) and \( \nu \). The latter step can be done without loss of generality [47].

Using \( \Pi_E \Pi_{E'} = \delta_{E,E'} \Pi_E \), the trace over the bath degrees of freedom of Eq. (A1) yields

\[
\sum_E \partial_t \hat{\rho}_S(E, t) = - \frac{\sum_{E_\alpha, E_\gamma} \int_0^t dt \left[ \langle B_\alpha(t) B_\gamma(s) \rangle_E \rho_{\alpha}(t) \rho_{\gamma}(s) \right] \hat{\rho}_S(E, t) \rho_{\gamma}(s) \rho_{\alpha}(t)}{\sum_{E_\alpha, E_\gamma} \int_0^t dt \left( \langle B_\alpha(s) B_\gamma(t) \rangle_E \rho_{\alpha}(s) \rho_{\gamma}(t) \right) \rho_{\gamma}(s) \rho_{\alpha}(t)}.
\]

(A3)

Here, we defined the microcanonical average \( \langle \ldots \rangle_E \equiv \text{tr}_B \{ \ldots \rho_{\text{mic}}(E) \} \). To obtain an equation for \( \rho_S(E, t) \), we drop the sum over \( E \), which appears on all sides. Then, after coming back to the Schrödinger picture and after a change of integration variables \( \tau = t - s \), we are left with

\[
\partial_t \rho_S(E, t) = -i[H_S, \rho_S(E, t)] - \sum_{E_\alpha, E_\gamma} \int_0^t dt \left[ \langle B_\alpha(\tau) B_\gamma(s) \rangle_E \rho_{\alpha}(t) \right] \rho_{\gamma}(s) \rho_{\alpha}(t) \rho_{\gamma}(s) \rho_{\alpha}(t) + \rho_{\gamma}(s) \rho_{\alpha}(t) \rho_{\gamma}(s) \rho_{\alpha}(t)
\]

(A4)

This is the microcanonical Redfield master equation, which only relies on the Born and weak-coupling approximation. As such, it is still quite general, but hard to deal with in practice and theory.

We next take a look at the correlation functions. Denoting the energy of a single bath eigenstate by \( E_i = E + \delta_i \), we obtain and approximate

\[
\langle B_\alpha(\tau) B_\gamma(s) \rangle_E = \sum_{E'} \frac{e^{i(E-E')\tau}}{V_E} \sum_{E' \in (E - \delta, E]} \sum_{E' \in (E' - \delta, E']} \text{tr}_B \{ \Pi_{E_\alpha} \Pi_{E_\gamma} \} e^{i(\delta_i - \delta_i')\tau} \approx \sum_{E'} \frac{e^{i(E-E')\tau}}{V_E} \text{tr}_B \{ \Pi_{E_\alpha} \Pi_{E_\gamma} \} \equiv \sum_{E'} \frac{e^{i(E-E')\tau}}{V_E} f_{\alpha(\gamma,E',E')}.
\]

(A5)

This approximation is similar but not identical to the one of Sec. II B. We expect it to be valid in the limit where \( \delta \) is small compared to differences in the eigenspectrum of \( H_S \) and if the bath correlation functions are peaked around \( \tau = 0 \), i.e., in the limit typically associated with Markovianity. For later purposes we also note the symmetries

\[
f_{\alpha(\gamma,E',E')} = f_{\gamma(\alpha,E',E')}, \quad f_{\alpha(\gamma,E',E')} = f_{\gamma(\alpha,E',E')} \in \mathbb{C}.
\]

(A6)

In this Markovian limit we then send the integration limit \( t \) to infinity and we are left with

\[
\partial_t \rho_S(E, t) = -i[H_S, \rho_S(E, t)]
\]

\[
- \sum_{E'} \sum_{E_\alpha, E_\gamma} \sum_{s,s'} S^{ss'}_{\gamma} \int_0^\infty dt e^{i(E-E'-\epsilon_s+\epsilon_{s'})\tau} f_{\alpha(\gamma,E',E')} \left\{ \frac{1}{V_{E'}} S_{\alpha}|s\rangle \langle s'\rho_S(E, t) - \frac{1}{V_{E'}} S_{\alpha} \rho_S(E, t) |s\rangle \langle s'| \right\}
\]

\[
- \sum_{E'} \sum_{E_\alpha, E_\gamma} \sum_{s,s'} S^{ss'}_{\gamma} \int_0^\infty dt e^{i(E-E'-\epsilon_s+\epsilon_{s'})\tau} f_{\alpha(\gamma,E',E')} \left\{ \frac{1}{V_{E'}} \rho_S(E, t) |s\rangle \langle s'| S_{\alpha} - \frac{1}{V_{E'}} |s\rangle \langle s'| \rho_S(E, t) S_{\alpha} \right\},
\]

(A7)

where we decomposed \( S_{\gamma}(\tau) = \sum_{s,s'} S^{ss'}_{\gamma} e^{i(\epsilon_s - \epsilon_{s'})\tau} |s\rangle \langle s'| \) in the (assumed to be non-degenerate) energy eigenbasis of \( H_S \). Next, we use \( \int_0^\infty dt e^{ixt} = \pi \delta(x) \), where we neglected any imaginary (Lamb shift) contributions. This allows us
to write
\[ \partial_t \rho_S(E, t) = -i[H_S, \rho_S(E, t)] \]
\[ - \sum_{\alpha, \gamma} \sum_{s, s'} \frac{\pi S_{ss'}^{s_s} S_{ss'}^{s_s'}}{V_E} f_{\alpha \gamma}(E, E + \epsilon_s - \epsilon_{s'}) \left\{ \rho_S(E, t) \langle s' | \rho_S(E, t) - \frac{V_E}{V_E + \epsilon_s - \epsilon_{s'}} | s \rangle \langle s | \rho_S(E, t + \epsilon_s - \epsilon_{s'}) \rangle \right\} \]
\[ - \sum_{\alpha, \gamma} \sum_{s, s'} \frac{\pi S_{ss'}^{s_s} S_{ss'}^{s_s'}}{V_E} f_{\gamma \alpha}(E, E - \epsilon_s + \epsilon_{s'}) \left\{ \rho_S(E, t) | s \rangle \langle s | \rho_S(E, t + \epsilon_s + \epsilon_{s'}) \rangle \right\} . \] (A8)

Finally, we apply the secular approximation assuming also that the differences in the energy spectrum of \( H_S \) are non-degenerate. This amounts to replacing
\[ \sum_{s, s'} S_{s_s}^{s_{s_s}} S_{s_s}^{s_{s_s}} | s \rangle \langle s' | \rho \to \sum_{s, s'} S_{s_s}^{s_{s_s}} S_{s_s}^{s_{s_s}} | s \rangle \langle s' | \rho, \]
\[ \sum_{s, s'} S_{s_s}^{s_{s_s}} S_{s_s}^{s_{s_s}} | s \rangle \langle s' | \rho | s \rangle \langle s | \to \sum_{s, s'} S_{s_s}^{s_{s_s}} S_{s_s}^{s_{s_s}} | s \rangle \langle s' | \rho | s \rangle \langle s | . \] (A9)

Hence, we end up with the microcanonical Born-Markov secular master equation:
\[ \partial_t \rho_S(E, t) = -i[H_S, \rho_S(E, t)] \]
\[ - \sum_{\alpha, \gamma} \sum_{s, s'} \frac{\pi S_{ss'}^{s_s} S_{ss'}^{s_s'}}{V_E} f_{\alpha \gamma}(E, E - \epsilon_s + \epsilon_{s'}) \left\{ \rho_S(E, t) \langle s' | \rho_S(E, t) - \frac{V_E}{V_E + \epsilon_s + \epsilon_{s'}} | s \rangle \langle s | \rho_S(E, t + \epsilon_s + \epsilon_{s'}) \rangle \right\} \]
\[ - \sum_{\alpha, \gamma} \sum_{s, s'} \frac{\pi S_{ss'}^{s_s} S_{ss'}^{s_s'}}{V_E} f_{\gamma \alpha}(E, E + \epsilon_s - \epsilon_{s'}) \left\{ \rho_S(E, t) \langle s | \rho_S(E, t + \epsilon_s - \epsilon_{s'}) \rangle \right\} . \] (A10)

It is well-known that all quantum coherences exponentially die out with time and that the dynamics of this MME is well-captured by a Pauli-like master equation. Defining \( p_{sE}(t) \equiv \langle s | \rho_S(E, t) | s \rangle \), we end up with the Pauli MME (43) as stated in the main text.