Equality statements for entropy change in open systems

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Abstract

The entropy change of a (non-equilibrium) Markovian ensemble is calculated from (1) the ensemble phase density \( p(t) \) evolved as iterative map, \( p(t) = M(t)p(t-\Delta t) \) under detail balanced transition matrix \( M(t) \), and (2) the invariant phase density \( \pi(t) = M(t)^\infty \pi(t) \). A virtual measurement protocol is employed, where variational entropy is zero, generating exact expressions for irreversible entropy change in terms of the Jeffreys measure, \( \mathcal{J}(t) = \sum_\Gamma [p(t) - \pi(t)] \ln [p(t)/\pi(t)] \), and for reversible entropy change in terms of the Kullbach-Leibler measure, \( D_{KL}(t) = \sum_\Gamma \pi(0) \ln [\pi(0)/\pi(t)] \).

Five properties of \( \mathcal{J} \) are discussed, and Clausius’ theorem is derived.

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Reversible manipulation is the principal tool of the thermodynamicist. Reversibility appears in two forms: the quasi-static time-forward reversible transition and the microscopically-reversible time-reversed (or adjoint) stochastic transition. Application of microscopic reversibility to the path integral formulation of stochastic processes has resulted in a set of fluctuation theorems (FT) for systems arbitrarily far from equilibrium. FT, despite their elegance, do not provide a much needed general definition of entropy change—an equality statement providing the entropy change for any transition—of an ensemble of Markovian systems. Here, using both the quasi-static and time-reversed transitions in the path integral approach to the dynamics of a Markovian system, we produce these equality expressions for microscopic and macroscopic entropy change.

A collection of $M$ classical particles undergoing Hamiltonian dynamics is partitioned, through scale separation, into system and bath. The system, consisting of $N$ particles is transformed into a Markovian stochastic process described by $6N$ generalized coordinates. Phase space and time are taken as discrete quantities. Each coordinate is an $m$-tuple, and time consists of equally spaced intervals, $\Delta \tau = \tau_{i+1} - \tau_i$. The system trajectory is given as the time evolution of a phase point, $\sigma(\tau) = \delta \left[ \left( x; \tau \right) - \left( x_0; \tau \right) \right]$, in phase space $\Gamma$, a $(6N \times m)$-tuple, with $x, x_0 \in \Gamma$, according to the stochastic iterative map

$$\sigma(\tau_i) = M_{\tau_i} \sigma(\tau_{i-1}),$$  

(1)

where $M_{\tau_i} = M_{\tau_i}(\sigma(\tau_i) | \sigma(\tau_{i-1}))$ is interpreted as a stochastic matrix. Real systems, which operate under colored (OU) noise, are managed by requiring that the discrete time step in (1) be much longer than the correlation time of the noise. Following Gibbs, we consider an ensemble of such collections. Interpreting $M_{\tau_i}$ as a transition matrix (rather than a stochastic matrix) and defining the phase probability as the normalized density of phase points, $P(\tau_i) = \bar{\sigma}(\tau_i)$, the dynamics of the ensemble is a time-inhomogeneous Markov chain

$$P(\tau_i) = M_{\tau_i} P(\tau_{i-1}).$$  

(2)
It is assumed that $M_{\tau_i}$ is a known quantity obtained through experimental parametrization or on the basis of theory. From $M_{\tau_i}$ and given starting phase density $p(\tau_0) = P(\tau_0)$, two time-dependent quantities of interest are determined—the time-dependent phase density $p(\tau_i)$ and the time-dependent invariant phase density, $\pi(\tau_i) = M_{\tau_i}^\infty \pi(\tau_i)$, obtained as follows: the dynamics at time $\tau_i$ are stopped, then the density is evolved in virtual time $t_i \to \infty$ under stationary $M_{\tau_i}$, according to (2). For nonequilibrium system ensembles, $\pi$ is a virtual quantity. For equilibrium ensembles that undergo quasi-static perturbation, $\pi$ is a real quantity. Our results apply to ensembles that evolve according to (2) with transition matrices $M_{\tau_i}$ that are Hermitian. These systems possess three important properties [11]: (i) Microscopic reversibility—$M_{\tau_i}$ is self-adjoint [3, 4]: $M_{\tau_i} \pi(\tau_i) = \tilde{M}_{\tau_i} \pi(\tau_i)$. (ii) Invariance—the invariant distribution of a stationary Markov process is independent of the ensemble history: $\lim_{t_n \to \infty} M_{\tau_i}^{t_n} p(\tau_i) = \pi(\tau_i)$. (iii) Stationarity—with invariant density: $\pi(\tau_i) = M_{\tau_i} \pi(\tau_i)$. Microscopic reversibility is a property of physical systems [12] and a fundamental postulate of physics [2].

**General Perterbation**—With these properties in mind, we calculate the entropy change of a thermodynamic ensemble of Markovian systems undergoing an arbitrary forced perturbation. Using the prescription in Fig. 1, entropy change for the ensemble transition along the path increment $\gamma_i$ (bold arrows) is evaluated using three measurements that,
being virtual, do not perturb the system \[1, 2\]. The path \( \gamma \) taken by one system starting at \( \sigma(t_0) \) is given by the time-ordered collection of phase points visited by the system \( \{\sigma(t_0)\sigma(t_1) \cdots \sigma(t_n)\} \) under evolution by \( M_{t_0}M_{t_1} \cdots M_{t_n} \),

\[
\gamma \equiv \sigma(t_0) \xrightarrow{M_{t_1}} \sigma(t_1) \xrightarrow{M_{t_2}} \cdots \xrightarrow{M_{t_{n-1}}} \sigma(t_n).
\]

The adjoint path is

\[
\tilde{\gamma} \equiv \tilde{\sigma}(t_n) \xleftarrow{\tilde{M}_{t_n}} \tilde{\sigma}(t_{n-1}) \xleftarrow{\tilde{M}_{t_{n-1}}} \cdots \xleftarrow{\tilde{M}_{t_2}} \tilde{\sigma}(1) \xleftarrow{\tilde{M}_{t_1}} \tilde{\sigma}(0),
\]

where \( \tilde{M}_{t_j} = \tilde{M}_{t_j}(\tilde{\sigma}(t_j)\tilde{\sigma}(t_{j-1})) \). The adjoint path starts where \( \gamma \) ends, \( \tilde{\sigma}(t_0) = \sigma(t_n) \). For a system in state \( \sigma(t_0) \), the probability that it follows the path \( \gamma \) is given by the product of the single time step transition probabilities,

\[
P(\gamma|p) = \prod_{i=1}^n M_{t_i},
\]

where \( p = P(\sigma(t_0)) \). Similarly, for the adjoint transition, the conditional adjoint path probability is \( P(\tilde{\gamma}|\tilde{p}) = \prod_{i=1}^n \tilde{M}_{t_i} \), where \( \tilde{p} = P(\tilde{\sigma}(t_0)) \) is the probability that the system starts in state \( \tilde{\sigma}(t_0) \). Using the definition of conditional probability and taking the quotient of path probabilities, we obtain

\[
\frac{P(\gamma)}{P(\tilde{\gamma})} = \frac{P(\gamma|p)}{P(\tilde{\gamma}|\tilde{p})} = \prod_{i=1}^n \frac{M_{t_i}}{\tilde{M}_{t_i}} \frac{p}{\tilde{p}}. \tag{3}
\]

The conditional path probability is also given as a function of action \( [1, 4] \), \( P(\gamma|p) = \exp(-\sum_i A_{t_i}(\gamma)) \), and we note the correspondence, \( \ln M_{t_i} = -A_{t_i} \). Defining the microscopic entropy change of the collection \( [4, 10] \)

\[
\tilde{\delta}S \equiv \ln P(\gamma) - \ln P(\tilde{\gamma})
\]

\[
= \ln \left[ \frac{p}{\tilde{p}} \exp(\sum_i \ln M_{t_i} - \ln \tilde{M}_{t_i}) \right], \tag{4}
\]

we obtain a microscopic entropy balance equation

\[
\tilde{\delta}S = \Delta S_\gamma + \ln \left[ \frac{p}{\tilde{p}} \right], \tag{5}
\]

involving gain of entropy by the heat bath, \( \Delta S_\gamma = \sum_i \ln M_{t_i}/\tilde{M}_{t_i} \), and gain of entropy by the system, \( \ln(p/\tilde{p}) \). In \( [4] \), entropy and action are on equal footing: entropy is proportional to the logarithm of exponentiated action. Evaluation of \( [5] \) is straightforward when some
path $\gamma$ can be identified where $\Delta S = 0$. We show that the measurement in Fig. 1 is along such a path.

Each path $\gamma^a_i$ involves the evolution in virtual time $t_0, t_1, \ldots, t_n$ of the ensemble starting in $p(\tau_{i-1})$ to the stationary distribution $\pi(\tau_{i-1})$. The evolution is a virtual time-homogeneous transition governed by the transition matrix $M_{\tau_{i-1}}$. For each $\gamma^a_i$, (5) provides, $\Delta S_{\gamma^a_i} = \delta S_{\gamma^a_i} - \ln [p(\tau_{i-1})/\pi(\tau_{i-1})]$. Using the property of invariance (ii), the path probability density is

$$P(\gamma^a_i) = M_{\tau_{i-1}}^\infty p(\tau_{i-1}) = \pi(\tau_{i-1}).$$

Application of detailed balance (i) followed by sequential application of stationarity (iii) to the adjoint path probability density yields

$$P(\tilde{\gamma}^a_i) = \tilde{\delta S}_{\gamma^a_i} = \ln \left[ \frac{P(\gamma^a_i)}{P(\tilde{\gamma}^a_i)} \right] = 0,$$

yielding, $\Delta S_{\gamma^a_i} = -\ln \left[ \frac{\pi(\tau_{i-1})}{\pi(\tau_{i})} \right]$. The macroscopic entropy over the disjoint paths $\gamma^a_i$ is the ensemble averaged entropy

$$\Delta S_{\gamma^a} = -\sum_{i=0}^{n-1} \langle \ln \left[ \frac{p(\tau_i)}{\pi(\tau_i)} \right] \rangle_{p(\tau_i)} ,$$

where $\langle f(\Gamma) \rangle_{p(\Gamma)} = \sum_{\Gamma} p(\Gamma) f(\Gamma)$.

Each path $\gamma^b_i$ involves the virtual evolution of the invariant starting distribution $\pi(\tau_{i-1})$ to the invariant distribution $\pi(\tau_i)$ under (virtual) time-homogeneous evolution by $M_{\tau_i}$. For each $\gamma^b_i$, (5) provides, $\Delta S_{\gamma^b_i} = \delta S_{\gamma^b_i} - \ln [p(\tau_{i-1})/\pi(\tau_i)]$. By the same arguments used in (5) and (7), for each path $\gamma^b_i$, \(\tilde{\delta S}_{\gamma^b_i} = \ln \left[ \frac{P(\gamma^b_i)}{P(\tilde{\gamma}^b_i)} \right] \) = 0, yielding, $\Delta S_{\gamma^b_i} = -\ln \left[ \frac{\pi(\tau_{i-1})}{\pi(\tau_{i})} \right]$. We concatenate the $\gamma^b_i$ path segments into a continuous virtual path $\gamma^b$ for the evolution of $\pi$. The microscopic entropy over the thermodynamically reversible path $\gamma^b$ is, after cancelling terms, $\Delta S_{\gamma^b} = \sum_{i=1}^n \Delta S_{\gamma^b_i} = -\ln \left[ \frac{\pi(\tau_0)}{\pi(\tau_n)} \right]$. The
reversible macroscopic entropy flow \( \text{into the system} \) during \( \gamma^b \) is

\[
\Delta S_{\text{rev}} = -\Delta S_{\gamma^b} = \langle \ln \frac{\pi(\tau_0)}{\pi(\tau_n)} \rangle_{\pi(\tau_n)},
\]

which is the relative, or Kullbach-Leibler, entropy \( D_{KL}[x, y] = \sum_{x} x \ln(x/y) \). \( \Box \)

Each path \( \gamma^c_i \) involves the adjoint (virtual) time-homogeneous evolution of the ensemble starting from the invariant distribution \( \pi(\tau_i) \) to the real distribution \( p(\tau_i) \) under \( \overline{M}_{\tau_i} \). For each \( \gamma^c_i \) the entropy \( \Delta S_{\gamma^c_i} = \delta S_{\gamma^c_i} - \ln [\pi(\tau_i)/p(\tau_i)] \). Again, by the same arguments used in (6) and (7), for each path \( \gamma^c_i \), \( \delta S_{\gamma^c_i} = \ln [\mathcal{P}(\gamma^c_i)/\mathcal{P}(\overline{\gamma}^c_i)] = 0 \), yielding, \( \Delta S_{\gamma^c_i} = -\ln [\pi(\tau_i)/p(\tau_i)] \). The macroscopic entropy change over the disjoint paths \( \gamma^c_i \) is

\[
\Delta S_{\gamma^c} = -\sum_{i=1}^{n} \langle \ln \frac{\pi(\tau_i)}{p(\tau_i)} \rangle_{\pi(\tau_i)}. \quad (10)
\]

From (8) and (10), the irreversible macroscopic entropy flow \( \text{into the system} \) over \( \gamma^a \) and \( \gamma^c \), \( \Delta S_{\text{irrev}} = -(\Delta S_{\gamma^a} + \Delta S_{\gamma^c}) \), is

\[
\Delta S_{\text{irrev}} = B_0 + \sum_{i=1}^{n-1} \langle \ln \frac{p(\tau_i)}{\pi(\tau_i)} \rangle_{p(\tau_i)} - \pi(\tau_i) + B_n, \quad (11)
\]

where \( B_0 = \sum_{\tau} p(\tau_0) \ln p(\tau_0)/\pi(\tau_0) \) and \( B_n = \sum_{\tau} \pi(\tau_n) \ln \pi(\tau_n)/p(\tau_n) \) are boundary Kullbach-Leibler integrals. The sum in (11) is over the Jeffreys invariant divergence measure, \( J[x, y] = \sum_{\Gamma} (x - y) \ln(x/y) \). Jeffreys \( \Box \) and others \( \Box \) have commented on the many remarkable properties of \( J \).

**Perturbation \#1**—We apply the results obtained for the general perturbation to two specific, and important, perturbations (Fig. 2). In Fig. 2a, the system is perturbed from one equilibrium state to another. For the (virtual) equilibrium path (dashed line), macroscopic entropy flow into the system is, from (9), the Kullbach-Leibler entropy, \( \Delta S_{\text{rev}} = D_{KL}[\pi(\tau_0), \pi(\tau_n)] \). For the (real) non-equilibrium path (solid line), macroscopic entropy is the sum of the reversible entropy (9) and the irreversible entropy (11), \( \Delta S_{\text{tot}} = \Delta S_{\text{rev}} + \Delta S_{\text{irrev}} \). Here, \( p(\tau_0) = \pi(\tau_0) \) and \( p(\tau_n) = \pi(\tau_n) \), causing \( B_0 \) and \( B_n \) to vanish. The irreversible entropy flow into the system is the discrete time integral over the
Jeffreys invariant measure, $\Delta S_{\text{irrev}} = \sum_{i=1}^{n-1} J[p(\tau_i), \pi(\tau_i)]$. The total entropy flow into the system is

$$\Delta S_{\text{tot}} = D_{KL}[\pi(\tau_0), \pi(\tau_n)] + \sum_{i=1}^{n-1} J[p(\tau_i), \pi(\tau_i)].$$

**Perturbation #2**—In Fig. 2b, we consider an ensemble of systems undergoing periodic perturbation with period $\omega \Delta \tau$, where $M_{\tau_i} = M_{\tau_i + \omega}$. Using (2), an initial phase density $p(\tau_0) = P(\sigma(\tau_0))$, is prepared from some arbitrary phase density $p(\tau_{-\Omega})$ through the equilibration process: $p(\tau_0) = (M_{\tau_n} M_{\tau_{n-1}} \ldots M_{\tau_1})^{\Omega/\omega} p(\tau_{-\Omega})$, where $\Omega/\omega \in \mathbb{I}^+ \gg 1$. While the Poincaré recurrence time for any one system may be extremely long, the recurrence time for the ensemble is $\omega \Delta \tau$. From (9) and using the property, $\pi(\tau_0) = \pi(\tau_n)$, we obtain $\Delta S_{\text{rev}} = D_{KL}[\pi(\tau_0), \pi(\tau_n)] = 0$. From (11) and the property, $p(\tau_0) = p(\tau_n)$, we obtain $\Delta S_{\text{irrev}} = \sum_{i=1}^{n} J[p(\tau_i), \pi(\tau_i)]$. The total entropy flow into the system over one cycle of
perturbation is
\[ \Delta S_{\text{tot}} = \sum_{i=1}^{n} J[p(\tau_i), \pi(\tau_i)]. \]  
(13)

The total entropy transferred from the bath to the system over a thermodynamic cycle is the time integral of the Jeffreys divergence between real and invariant phase densities.

Properties of \( J \)—The properties of \( J \) generate some important conclusions. \( J \) is almost positive definite, meaning (i) \( J[x, y] \geq 0 \) and (ii) \( J[x, x] = 0 \) only when \( x = y \). (iii) \( J \) is symmetric: \( J[x, y] = J[y, x] \). (iv) \( J \) is a linear measure: \( \det \left( \partial^2 J / \partial x_i \partial x_j \right) = 0 \).

\( D_{KL} \) satisfies (i), (ii) [20] and (iv). See EPAPS Document No. [ ] for proofs and further discussion.

For an isothermal (cannonical) system, \( \Delta S = -\Delta S_{\text{tot}} \), is the heat flow per temperature from the system to the bath during a periodic perturbation, \( \Delta S = \oint \beta dQ \), where \( \beta = 1/k_B T \) is inverse temperature in units of energy. Clausius’ statement of the second law of thermodynamics is obtained from (13) and properties (i) and (ii),

\[ \oint \beta dQ \leq 0. \]

Practical application of (13) to many-body systems derives from a fifth property of \( J \), (v) decomposability [19]: for a system with a decomposable Markov transition matrix

\[
\mathbb{M} = \begin{bmatrix}
A & 0 \\
0 & \mathbb{B}
\end{bmatrix},
\]

the phase density decomposes: \( p = p_{A \mathbb{B}} \), \( \pi = \pi_A \pi_B \), phase space decomposes: \( \Gamma = \Gamma_A \Gamma_B \), and the invariant measure decomposes: \( J[p, \pi] = J^A[p_A, \pi_A] + J^B[p_B, \pi_B] \). Defining, \( \Delta S^k = \sum_i J^k[p_i, \pi_i] \), and using (v), we find that macroscopic entropy is extensive, \( \Delta S = \Delta S^A + \Delta S^B \). The most immediate application of decomposability is the overdamped system where \( \Gamma_B \) comprises momentum space and momentum is always equilibrated, \( p_B = \pi_B \).

Using (13) and properties (ii) and (v), we obtain, \( \Delta S = \Delta S_A \). Further reduction may be possible with a suitable choice of basis for \( \Gamma_A \) [21] and also upon coarse graining of the system [7, 22].
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A. Some comments

Aside from generating equality statements for reversible and irreversible entropy change, perhaps our most important contribution is providing a purely virtual integration protocol for evaluating a variation. Being virtual, the integration can not introduce uncertainty into the system [1, 2]. This measurement protocol is likely to find application beyond statistical mechanics.

We note that Gibbs entropy, $S = -\ln p$, and Boltzmann entropy, $S = -\ln \pi$, follow as boundary terms from the definition of variational entropy $\delta S$ when $p$ or $\tilde{p}$ are evaluated at $\pi$.

B. Some properties of the Jeffreys divergence $J$ and the Kullback-Leibler divergence $D$

The Jeffreys divergence measure is defined

$$J[x, y] = \sum_{\Gamma} (x - y) \ln(x/y).$$

The Kullback-Leibler divergence measure is defined

$$D[x, y] = \sum_{\Gamma} x \ln(x/y).$$

**Theorem 1.** $J$ is almost positive definite, meaning (i) $J[x, y] \geq 0$ and (ii) $J[x, y] = 0$ only when $x = y$.

**Proof** We consider, element-wise, the probability densities $x = \{x\}$, $y = \{y\}$, and the Jeffreys measure, $J[x, y] = \sum_{\Gamma} J_i[x, y]$. For $x > y$, $(x - y) > 0$ and $\ln(x/y) > 0$; therefore, $J_i[x, y] = (x - y) \ln(x/y) > 0$. For $x < y$, $(x - y) < 0$ and $\ln(x/y) < 0$; therefore, $J_i[x, y] > 0$. For $x = y$, $(x - y) = 0$ and $\ln(x/y) = 0$; therefore, $J_i[x, y] = 0$. 

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Theorem .2 $\mathcal{J}$ is symmetric: $\mathcal{J}[x,y] = \mathcal{J}[y,x]$ 

Proof $\mathcal{J}$ is evaluated element-wise. Both terms $(x-y)$ and $\ln(x/y)$ are odd under exchange of $x$ and $y$. The product of two odd functions is even.

Theorem .3 $\mathcal{J}$ does not satisfy the triangle inequality: $\mathcal{J}[x, z] \leq \mathcal{J}[x, y] + \mathcal{J}[y, z]$.

Proof The proof is by example (Nikolai Chernov, personal communication). Let $x = [0.25, 0.75]$, $y = [0.50, 0.50]$, and $z = [0.75, 0.25]$. $\mathcal{J}[x, y] = 0.27$; $\mathcal{J}[y, z] = 0.27$; $\mathcal{J}[x, z] = 1.10$. We obtain, $\mathcal{J}[x, z] > \mathcal{J}[x, y] + \mathcal{J}[y, z]$. By not satisfying the triangle equality, the Jeffreys measure falls short of being a topologic metric. For this reason, the term “Jeffreys divergence measure” is the preferred over the “Jeffreys distance measure.”

Theorem .4 $\mathcal{J}$ is a linear measure.

Proof The Hession of $\mathcal{J}[x, y]$, 

$$H(\mathcal{J}[x, y]) = \begin{bmatrix} \frac{\partial^2 \mathcal{J}}{\partial x \partial x} & \frac{\partial^2 \mathcal{J}}{\partial x \partial y} \\ \frac{\partial^2 \mathcal{J}}{\partial y \partial x} & \frac{\partial^2 \mathcal{J}}{\partial y \partial y} \end{bmatrix},$$

is evaluated: $\frac{\partial^2 \mathcal{J}}{\partial x \partial x} = (x + y)/x^2$, $\frac{\partial^2 \mathcal{J}}{\partial y \partial y} = (x + y)/y^2$, and $\frac{\partial^2 \mathcal{J}}{\partial x \partial y} = \frac{\partial^2 \mathcal{J}}{\partial y \partial x} = -(x + y)/xy$. By substitution we find, $\det H(\mathcal{J}) = 0$.

Theorem .5 $\mathcal{D}$ is a linear measure.

Proof The Hession of $\mathcal{D}[x, y]$ is evaluated: $\frac{\partial^2 \mathcal{D}}{\partial x \partial x} = 1/x$, $\frac{\partial^2 \mathcal{D}}{\partial y \partial y} = x/y^2$, and $\frac{\partial^2 \mathcal{D}}{\partial x \partial y} = \frac{\partial^2 \mathcal{D}}{\partial y \partial x} = -1/y$. By substitution we find, $\det H(\mathcal{D}) = 0$.

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